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Sediment and Radionuclide Transport in Rivers

Phase I Field Sampling Program During Mean Flow Cattaraugus and Buttermilk Creeks, New York

R. M. Ecker
Y. Onishi

August 1979

Prepared for
the U.S. Nuclear Regulatory Commission

Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
by Battelle Memorial Institute

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SEDIMENT AND RADIONUCLIDE TRANSPORT IN RIVERS

PHASE 1 - FIELD SAMPLING PROGRAM DURING MEAN FLOW CATTARAUGUS AND BUTTERMILK CREEKS, NEW YORK

R. M. Ecker
Y. Onishi

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Pacific Northwest Laboratory
Richland, Washington 99352

ABSTRACT

A field sampling program was conducted on Cattaraugus and Buttermilk Creeks, New York during November and December 1977 to investigate the transport of radionuclides in surface waters as part of a continuing program to provide data for application and verification of Pacific Northwest Laboratory's (PNL) sediment and radionuclide transport model, SERATRA. Suspended sediment, bed sediment and water samples were collected during mean flow conditions over a 45 mile reach of stream channel. Radiological analysis of these samples included primarily gamma ray emitters; however, some plutonium, strontium, curium and tritium analyses were also included. The principal gamma emitter found during the sampling program was ^{137}Cs where, in some cases, levels associated with the sand and clay size fractions of bed sediment exceeded 100 pCi/g. Elevated levels of ^{137}Cs and ^{90}Sr were found downstream of the Nuclear Fuel Services Center, an inactive plutonium reprocessing plant and low level nuclear waste disposal site. Based on radionuclide levels in upstream control stations, ^{137}Cs was the only radionuclide whose levels in the creeks downstream of the site could confidently be attributed to the site during this sampling program. This field sampling effort is the first of a three phase program to collect data during low, medium and high flow conditions.

SUMMARY

One mechanism affecting the dispersal of radioactive materials in water bodies is radionuclide adsorption by sediment. Consequently, sediment transport is a major factor to consider when evaluating radionuclide migration. In rivers, sorption is important because radionuclides may be concentrated in the stream beds.

As part of a study on sediment and radionuclide transport in rivers, Pacific Northwest Laboratory (PNL) is investigating the effect of sediment on the transport of radionuclides in Cattaraugus and Buttermilk Creeks, New York, during different flow conditions. One source of radioactivity in these creeks is the Western New York Nuclear Service Center which consists of a low-level waste disposal site and a nuclear fuel reprocessing plant. Reprocessing operations were terminated in 1972 and waste disposal was discontinued in 1975. Other sources of radioactivity include fallout from worldwide weapons testing and natural background radioactivity.

The major objective of the PNL Field Sampling Program is to provide data on sediment and radionuclide characteristics in Cattaraugus and Buttermilk Creeks to verify the use of the Sediment and Radionuclide Transport model, SERATRA, for nontidal rivers. The sampling program has been divided into three phases: Phase 1, medium-flow condition; Phase 2, low-flow condition; and Phase 3, high-flow condition. This report covers the results of field sampling during medium-flow conditions of 30 November to 5 December 1977.

Suspended sediment, bed sediment and water samples were collected at ten transects covering approximately 45 miles of stream channel of Cattaraugus and Buttermilk Creeks. Radiological analysis of sand, silt and clay size fractions of suspended and bed sediment, and water were performed. Results of these analysis indicate that the principal radionuclides occurring in these two water courses, with levels higher than background levels, during the Phase 1 sampling program were ^{137}Cs and ^{90}Sr. Cesium-137 levels in bed sediments, exceeding 100 pCi/g, were found at the mouth of Cattaraugus Creek where it enters Lake Erie, and at some locations on Buttermilk Creek.

Cesium-137 levels in suspended sediments were highest in Buttermilk Creek and Franks Creek, a small tributary of Buttermilk Creek draining the Western New York Nuclear Service Center. In these suspended sediment samples ^{137}Cs levels greater than 10 pCi/g but less than 15 pCi/g were found. Cesium-137 levels in water of Cattaraugus and Buttermilk creeks were low and in most cases indistinguishable from background ^{137}Cs levels in water.

Detectable levels of ^{90}Sr, greater than background levels, were found in the suspended sediment samples of Buttermilk and Cattaraugus Creeks but could not be attributed to the NFS facility because of low levels found in Franks Creek, just downstream of the facility. One suspended sediment sample in Springville Reservoir had a ^{90}Sr value of 10.1 pCi/g. Strontium-90 levels in bed sediment and water downstream of the Nuclear Service Center were indistinguishable from background levels.

Concentrations of ^{134}Cs in bed and suspended sediments of Franks Creek and Buttermilk Creek were, in most cases, higher than levels measured at upstream control stations.

ACKNOWLEDGMENTS

This report summarizes the results of research conducted by the Battelle Memorial Institute's Pacific Northwest Laboratory for the U.S. Nuclear Regulatory Commission on Cattaraugus and Buttermilk Creeks, New York, during average flow conditions of November and December 1977. In addition to the authors of this report, significant contributions were by R. G. Parkhurst for his field and laboratory expertise; F. L. Thompson and S. M. Brown for their efforts in the field; and, S. J. Phillips, A. J. Scott and R. J. Serne for their scuba diving capabilities. The authors also wish to acknowledge the guidance and assistance provided by Dr. Phillip R. Reed of the U.S. Nuclear Regulatory Commission (NRC).

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INTRODUCTION

Radionuclides found in Cattaraugus and Buttermilk Creeks, New York, have originated in part from operation of the Western New York Nuclear Service Center at West Valley, New York, located about 30 miles south of Buffalo, New York. The Center is comprised of a low-level radioactive waste disposal area and a facility for the reprocessing of nuclear fuel. Nuclear Fuel Services, Inc. (NFS), the reprocessing plant operator, was licensed to operate the disposal site in November 1963. The nuclear fuel reprocessing operations were discontinued in 1972 and burial of wastes was suspended in March 1975.

Radioactive waste burial at West Valley has taken place in 25-ft-deep trenches. Rising water levels in the trenches during radioactive waste disposal operations became a problem in 1968 due to seepage through the trench caps. Radioactive water was pumped from the trenches to a burial site lagoon. Radioactivity was monitored in the lagoon and water was released to Franks Creek (Erdmans Brook) which flows into Buttermilk Creek about 0.5 miles below the site. In November 1972, radioactive water from the burial site lagoon was pumped to the NFS main plant lagoon No. 1 where it was treated in the low-level waste treatment plant prior to being released to the environment. In 1976, rising trench waters from two trenches had broken through the soil cover over the trenches.

Radioactivity in Cattaraugus and Buttermilk Creeks could have originated from several sources: directly from the burial trenches by overflow or seepage of trench water; indirectly by pumping to the burial site lagoon and later from the low-level treatment plant, and by erosion of soil and fill contaminated during burial operations; from airborne particulate matter from reprocessing plant stacks; from fallout due to weapons testing; and from natural background radioactivity.

Once the radionuclides have reached Buttermilk and Cattaraugus Creeks they are dispersed by mixing with the stream water or adsorbed from solution onto sediments. Sorption by sediments is an important mechanism affecting the migration of radionuclides throughout a water body because radioactive materials discharged can eventually accumulate in the stream bed.

The purpose of the work described in this report is to investigate the importance of sediment in the transport of radionuclides in Cattaraugus and Buttermilk Creeks, New York, during medium-flow conditions. The Phase 1 sampling program is the first part of a three-phase program to investigate the transport of sediment and radionuclides during medium, low, and high flows on Buttermilk and Cattaraugus Creeks. The primary objective of the overall program is to provide the necessary field data on flow characteristics, and sediment and radionuclide transport characteristics in Cattaraugus and Buttermilk Creeks for verification of the Pacific Northwest Laboratory's Sediment and Radionuclide Tranpsort model, SERATRA, in non-tidal rivers. The modeling effort is being accomplished under a separate NRC program (NRC FIN No. 2294).

The SERATRA model is a transport code capable of simulating the movement of sediment and radionuclides by accounting for sediment deposition and erosion, and sediment adsorption and desorption of radionuclides. The model is an unsteady two-dimensional code in the longitudinal and vertical directions which uses the finite element method of solution. The code has previously been applied to the Columbia River(1) and Clinch River.(2) However, available field data on sediment and radionuclide transport from these two rivers were not of sufficient quantity or proper format for rigorous verification of the code. The portion of the code for which there is very little existing data for verification is the particulate (those adsorbed by sediment) phase of radionuclide transport. Not only does the SERATRA code simulate the movement of radionuclides in the dissolved phase and particulate phase, but the particulate phase simulation can also be broken down into transport of radionuclides by different sediment size fractions, e.g., sand, silt and clay.

This report discusses the results of the Phase 1 sampling program - medium-flow conditions, conducted in November 1977. Included in the report is a brief site description, summary of previous radiological monitoring in Cattaraugus and Buttermilk Creeks, sampling methods and procedures, results of the Phase 1 sampling program, and discussion of results and conclusions.

SITE DESCRIPTION

The Western New York Nuclear Service Center, shown in Figure 1, is located about 30 miles south of Buffalo, New York. The Center consists of a 3345-acre site in the north central portion of Cattaraugus County. The Center is situated along an elongated rolling plain with glaciated bedrock hills along the eastern, western and southern boundaries, and Buttermilk Valley along the northern boundary. All surface drainage of the Center discharges into Buttermilk Creek. At the northwest end of the property, Buttermilk Creek joins Cattaraugus Creek which flows in a westerly direction into Lake Erie, 39 miles away. Cattaraugus Creek flows in a general westerly direction through Zoar Valley, Gowanda, and Cattaraugus Indian Reservation and empties into Lake Erie, 27 miles southwest of Buffalo. It is 20 stream miles from the confluence of Buttermilk and Cattaraugus Creeks to Gowanda and another 19 miles to the mouth of Cattaraugus Creek at Lake Erie. Franks Creek (Erdmans Brook), a tributary of Buttermilk Creek, serves as a receptor for runoff from the Center's burial site.

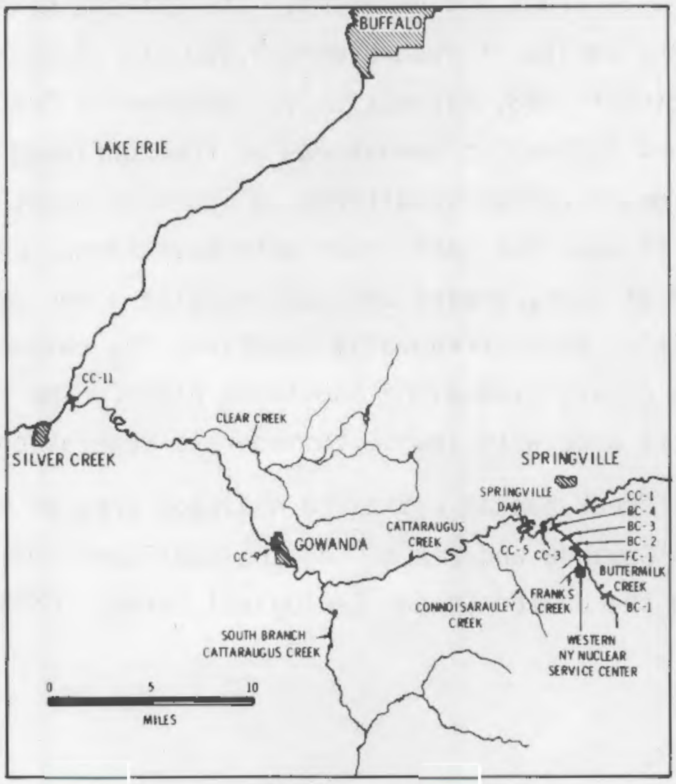

FIGURE 1. Map of Cattaraugus, Buttermilk and Franks Creeks

Three water courses, Franks Creek, Buttermilk Creek and Cattaraugus Creek, are the principal water courses of interest in the study of radionuclide transport in surface waters from the Western New York Nuclear Service Center. The following is a brief description of each of these water courses.

Franks Creek, commonly referred to as Erdmans Brook, includes the drainage area for both the low- and high-level nuclear burial sites. The creek flows into Buttermilk Creek about 0.5 miles downstream of the burial sites. The stream flow in the creek is intermittent, varying between 0 and 100 cubic feet per second (cfs). The creek is very narrow, varying in width from 2 to 10 ft. The creek is comprised of chutes and pools, and flows in some places through swampy areas. Stream gradients are moderately steep, and the creek shows active down-cutting through previously undisturbed glacial till which is comprised of a very stiff clayey material. This clayey material appears to be fairly resistant to erosion. The creek flows through a narrow and steep, V-shaped valley.

Buttermilk Creek has a drainage area of approximately 29.4 mi^2. For the period of record from October 1961 to September 1968, the average discharge of Buttermilk Creek was 46.5 cfs. The extreme maximum and minimum discharges during the period of record were 3,910 cfs on 28 September 1967 and 2.1 cfs on 10 October 1963, respectively. Buttermilk Creek flows into Cattaraugus Creek about 2.25 miles downstream of the confluence with Franks Creek. The creek width under normal conditions varies from about 20 ft at the upper end to about 75 ft near the confluence with Cattaraugus Creek. The channel bed is comprised of sand, gravel and cobbles with minor amounts of silt and clay size material. Water frequently overflows the channel banks leaving deposits of fine clayey silt along the flood plain. The flood plain varies from 300 to 500 ft wide with sparse to moderate vegetation.

Cattaraugus Creek has an estimated drainage area of 555 mi^2 at Lake Erie, 432 mi^2 at Gowanda and 218 mi^2 at the confluence with Buttermilk Creek. Based on the United States Geological Survey (USGS) flow data records for Cattaraugus Creek at Gowanda, New York, the average discharge for the period of record, 1940-1976, is 731 cfs. The extreme maximum and minimum daily discharges during the period of record were 34,600 cfs on 7 March 1956, and 6 cfs, respectively, on 21 August 1941.

Peak discharges generally occur on Cattaraugus Creek in October and November, prior to the onset of winter snowfall and again in February and March as a result of snowmelt. Low discharges generally occur during the summer months of July through September when rainfall is less and again during the winter months of December and January when persistent freezing conditions exist. Figure 2 is a summary of the 1975 and 1976 water year monthly discharge records of Cattaraugus Creek at Gowanda, New York. Cattaraugus Creek, as well as Buttermilk Creek, can be categorized as "flashy" due to their very rapid changes in discharge. Figure 3 is an excerpt from the 1976 water year discharge records showing the September daily discharges of Cattaraugus Creek at Gowanda. Discharges can be seen to vary by more than 5000 cfs in a period of one day.

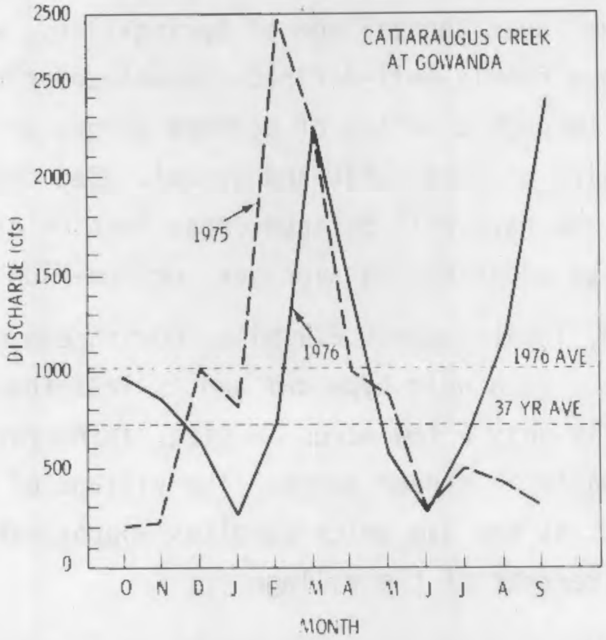

FIGURE 2. 1975-76 Monthly Discharges of Cattaraugus Creek at Gowanda

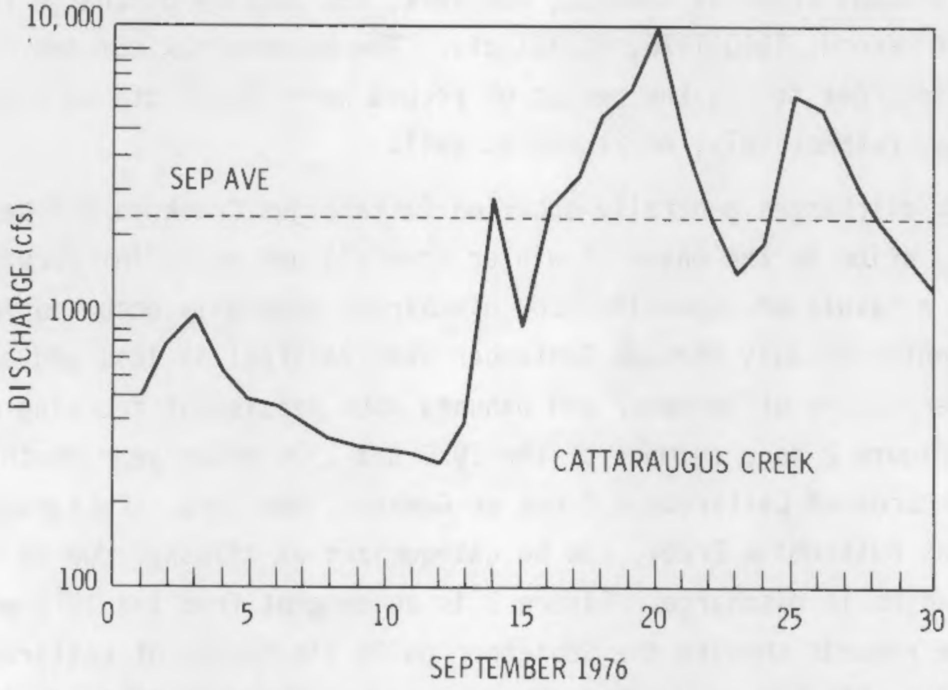

FIGURE 3. September Daily Discharges of Cattaraugus Creek at Gowanda

Cattaraugus Creek is generally free flowing except for a small impoundment (Springville Dam) near the Village of Springville. Water flow in the creek is confined to a fairly well-defined channel under normal discharge conditions and cuts through a series of bedrock gorges which are connected by shallow valley deposits of sand, silt and gravel. Bed deposits in the gorges appear to be slight and have well defined cross-section profiles. Little bank interaction appears to occur during average- or low-flow conditions.

Springville Dam, located about 2.5 miles downstream of the confluence with Buttermilk Creek, is a weir type dam and is less than 20 ft high. The pool behind the dam is only a few acres in size, is narrow and is surrounded by nearly vertical walls of a deep gorge. The Village of Springville operates a hydroelectric plant at the dam which supplies approximately 20% of the electric power requirements of the village.

PREVIOUS RADIOACTIVITY SURVEYS IN CATTARAUGUS AND BUTTERMILK CREEKS

Numerous studies of radioactivity levels in the surface-water drainage area of the Western New York Nuclear Service Center have been conducted since 1965. The following paragraphs summarize this work.

NEW YORK STATE HEALTH DEPARTMENT - GENERAL SURVEILLANCE PROGRAM 1965-1967(3)

The New York State Health Department conducted a 3-year surveillance program before, during and after startup of the Nuclear Fuel Services Reprocessing Plant. Included in the program were water and stream-bed samples on Buttermilk Creek, upstream of the confluence with Franks Creek; Cattaraugus Creek, upstream of the confluence with Buttermilk Creek; Thomas Corners Bridge on Buttermilk Creek; and, on Cattaraugus Creek at Felton Bridge, Springville Dam, Gowanda and Irving Bridge. Some samples were also taken from the waste lagoons at the site. Radiological analysis included ^{90}Sr, gross β, ^{137}Cs, ^{106}Ru, ^{95}Zr-Nb, ^{60}Co and ^{134}Cs. A number of analyses for plutonium and uranium were also conducted. Tables 1, 2, and 3 are the reported analysis data for water and silt.(3)

Radionuclide levels in water samples of Buttermilk Creek at Thomas Corners Bridge (Table 1) were substantially above background levels in 1967. Average ^{90}Sr, ^{137}Cs, gross β and tritium levels were 335 pCi/ℓ, 112 pCi/ℓ, 970 pCi/ℓ and 320,430 pCi/ℓ, respectively. These radionuclide levels in water decreased by at least an order of magnitude at Springville Dam. Gross and tritium levels decreased further in Cattaraugus Creek water samples at Gowanda and Irving Bridge. These decreases, however, were slight when compared to the initial decreases encountered when Buttermilk Creek samples are first diluted with water from Cattaraugus Creek. Comparison of 1966 and 1967 average and maximum ^{90}Sr and gross β levels at Springville Dam indicates a substantial decrease in concentrations in 1967.

TABLE 1. Radioactivity in Water Samples (NYS Health Department)

	Concentration (pCi/ℓ)							
	1965		1966		1967			
Location	Sr-90	Gross β	Sr-90	Gross β	Sr -90	Cs-137	Gross β	H-3
Buttermilk Creek, Upstream								
Average	<3		<3	2	<3	<20	6	1,570
Maximum	<3		<3	3	<3	<20	13	2,900
Cattaraugus Creek, Upstream								
Average	<3		<3	2.5	<3	<20	5	1,860
Maximum	<3		<3	3	8	50	10	4,500
Buttermilk Creek, Thomas Corners Bridge								
Average	<3		<3[a]	5.8[a]	335	112	970	320,430
Maximum	<3		<3	15	1,305	506	4,668	1,452,000
Cattaraugus Creek, Springville Dam								
Average	<3		62	116	24	<20	95	30,730
Maximum	<3		265	1,387	53	77	315	183,170
Cattaraugus Creek, Gowanda								
Average	<3						52	21,510
Maximum							157	100,950
Cattaraugus Creek, Irving Bridge								
Average	<3						51	24,710
Maximum	<3						112	82,670

(a) Samples not taken during period of discharge from Nuclear Fuels Services for latter half of 1966.

Table 2 is a summary of New York State Health Department's 1966 and 1967 analyses of radioactivity in the stream bed. Maximum concentrations of ^{90}Sr, ^{137}Cs, ^{106}Ru, ^{95}Zr-Nb and ^{60}Co in Buttermilk Creek at Thomas Corners Bridge in 1966 were 81 pCi/g, 179 pCi/g, 589 pCi/g, 222 pCi/g, and 9.2 pCi/g, respectively. The samples furthest downstream reported in this survey were located at Springville Dam, where the radionuclide levels decreased substantially from levels found in Buttermilk Creek. The 1966 levels in Springville Dam bed sediment, however, were generally higher than at Felton Bridge which is located on Cattaraugus Creek, just downstream of the confluence with Buttermilk Creek. Strontium-90 levels in the Cattaraugus and Buttermilk Creeks bed sediment were less in 1967 than in 1966. Cesium-137 levels in Springville Dam bed sediment in 1967 had an average concentration of 23 pCi/g, and the maximum concentration recorded was 44.4 pCi/g. Average and maximum ^{134}Cs levels in Springville Dam bed sediment in 1967 were 3.9 pCi/g

TABLE 2. Radioactivity in Silt Samples (NYS Health Department)

Location	Concentration (pCi/g) 1966 Sr-90	1966 Cs-137	1966 Ru-106	1966 Zr-Nb 95	1966 Co-60	1967 Sr-90	1967 Cs-137	1967 Ru-106	1967 Co-60	1967 Cs-134
Buttermilk Creek, upstream										
Average	0.3[a]	0.2[a]	1.07[a]			0.1[a]	0.08			
Maximum						0.2				
Cattaraugus Creek, upstream										
Average	1.6[a]	0.2	1.4	0.15	0.1	0.24	0.06[a]			
Maximum		0.3	1.9	0.1	0.1	0.6				
Buttermilk Creek, Thomas Corners Bridge										
Average	81[a]	176	318	155	7.7	2.6				
Maximum		179	589	222	9.2	4.8				
Cattaraugus Creek, Felton Bridge										
Average	4.6[a]	7.4	25.2	8.1	0.5	0.3				
Maximum		8.3	28.3	9.5	0.6	0.6				
Cattaraugus Creek, Springville Dam										
Average	3.8[a]	9.0[a]	32[a]	10.7[a]	0.4[a]	2.3[b]	23.0[b]	2.9[b]	0.3[b]	3.9[b]
Maximum						9.9	44.4	3.5	0.5	7.6

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[a] Only one sample reported

[b] Taken from various locations in reservoir

and 7.6 pCi/g, respectively. Average ^{106}Ru and ^{60}Co concentrations in bed sediment samples from Springville Dam in 1967 were 2.9 pCi/g and 0.3 pCi/g, respectively.

Some uranium and plutonium samples from Cattaraugus Creek were analyzed (Table 3) in 1966 and 1967. Water samples from upstream of the confluence with Buttermilk Creek and at Springville Dam in 1966 indicated that levels of ^{238}Pu, ^{239}Pu and ^{235}U did not increase above background levels in Springville Reservoir. Uranium-234 and ^{238}U concentrations in water samples from Springville Reservoir were slightly above background levels in 1966. Maximum ^{238}Pu, ^{239}Pu, ^{234}U, ^{235}U and ^{238}U concentrations in water samples from Springville Reservoir in 1967 were 1.16 pCi/ℓ, 0.7 pCi/ℓ, 0.15 pCi/ℓ, 0.11 pCi/ℓ, and 0.15 pCi/ℓ, respectively. These were, in most cases, above the 1966 background levels. Plutonium and uranium concentrations in Springville Reservoir bed sediment were generally low and probably represent background levels.

TABLE 3. Plutonium and Uranium in Water and Silt Samples (NYS Health Department)

Location	Concentration in Water (pCi/ℓ)									
	1966					1967				
	Pu-238	Pu-239	U-234	U-235	U-238	Pu-238	Pu-239	U-234	U-235	U-238
Cattaraugus Creek, upstream										
Average	<0.03	<0.02	0.19	<0.04	0.05					
Maximum	0.06	<0.03	<0.33	<0.15	0.12					
Cattaraugus Creek, Springville Dam										
Average	0.03	<0.02	0.28	<0.05	0.20	<0.5	<0.4	0.15	0.07	0.12
Maximum	0.063	<0.02	0.44	<0.12	0.41	1.16	0.7	0.15	0.11	0.15
	Concentration in Silt (pCi/g)									
Cattaraugus Creek, Springville Dam						ND	0.03	0.63		0.69

NEW YORK STATE AQUATIC ECOSYSTEM SURVEY, 1965-1967(4)

An aquatic ecosystem survey associated with the Nuclear Fuels Service Center was conducted in 1965 through 1967. The purpose of the survey was to identify and evaluate natural indicators of environmental radioactivity above background levels. Water and bed sediment samples from Buttermilk and Cattaraugus Creeks were taken in 1966 and 1967 and analyzed for ^{106}Ru-Rh, ^{137}Cs, ^{134}Cs and ^{60}Co. Table 4 is a summary of the radioactivity analyses of water and bed samples.

Analysis of water samples from Thomas Corners Bridge on Buttermilk Creek and Springville Reservoir on Cattaraugus Creek in 1967 indicated an order of magnitude decrease in concentrations between these two streams. For example, the ^{137}Cs level at Thomas Corners Bridge was 100 pCi/ℓ and at Springville Dam it was 10 pCi/ℓ.

Concentrations of radionuclides in bed sediment in 1967 samples showed highest concentrations at Thomas Corners Bridge on Buttermilk Creek and steadily decreased in a downstream direction to Zoar Bridge on

TABLE 4. New York State Aquatic Ecosystem Survey

	Concentrtation in Water (pCi/ℓ)							
	1966				1967			
Location	Ru-Rh 106	Cs 137	Cs 134	Co-60	Ru-Rh 106	Cs 137	Cs 134	Co-60
Buttermilk Creek, upstream	0	5	2	0	40	3	0	1
Buttermilk Creek, Thomas Corners Bridge	0	1	6	0	1000	100	10	10
Cattaraugus Creek, Springville Dam	0	1	0	0	100	10	0	4
	Concentration in Bottom Sediment (pCi/g)							
Buttermilk Creek, upstream	6.7	0.43	0.32	0.11	0.82	0.07	0.05	0.005
Buttermilk Creek, Thomas Corners Bridge					226	115	120	8
Cattaraugus Creek, Felton Bridge					53.4	62	12.8	0.69
Cattaraugus Creek, Springville Dam					45.7	28.8	7.4	0.47
Cattaraugus Creek, Frye Bridge					36.7	24.1	4.5	0.36
Cattaraugus Creek, Zoar Bridge					15.1	5.3	1.7	0.12

Cattaraugus Creek. Concentrations of ^{106}Ru-Rh, ^{137}Cs, ^{90}Zr-Nb and ^{60}Co in bed sediment at Thomas Corners Bridge were 226 pCi/g, 115 pCi/g, 120 pCi/g and 8 pCi/g, respectively. These radionuclide levels decreased from 1 to 7% of these values at Zoar Bridge.

U.S. DEPARTMENT OF HEALTH, EDUCATION AND WELFARE 1969 SURVEY(5)

The Public Health Service conducted a survey in 1969 for the purpose of developing minimum and optimum requirements for environmental surveillance programs around nuclear fuel reprocessing plants; designating the radionuclides and environmental pathways of greatest dosimetric significance; and providing methods for estimating the radiation dose to the populations in the area of a facility. In June 1969, they analyzed water and suspended sediment samples on Buttermilk and Cattaraugus Creeks as far downstream as Springville Dam. Water samples were analyzed for ^{3}H, gross α, gross β, ^{106}Ru, ^{137}Cs, ^{134}Cs, ^{90}Sr, ^{125}Sb and ^{60}Co. Suspended sediment samples were analyzed for gross β, ^{106}Ru, ^{137}Cs, ^{134}Cs, and ^{90}Sr.

As with results of previous studies of radioactivity in water of Cattaraugus and Buttermilk Creeks, this survey indicated that dissolved radionuclide concentrations in June 1969 were an order of magnitude less in Cattaraugus Creek than in Buttermilk Creek. Water and suspended sediment analysis for June and November 1969 are shown in Tables 5 and 6. Except for ^{137}Cs and ^{125}Sb levels in water, dissolved radionuclide concentrations were significantly lower in November, indicating the dilution effect of dissolved levels during high flow conditions. Radionuclide concentrations in suspended sediment, however, showed the opposite trend; i.e., during November with the greater discharge conditions, gross β, ^{137}Cs, and ^{134}Cs concentrations in suspended sedment were higher, possibly due to higher suspended sediment concentrations during this period.

Uranium and plutonium analyses of water and suspended sediment were also conducted in June and November 1969. The results are shown in Table 7. Concentrations of ^{232}U, ^{234}U, ^{238}U, ^{238}Pu, and ^{239}Pu in Buttermilk Creek water in June 1969 were 3.4 pCi/ℓ, 1.4 pCi/ℓ, 0.22 pCi/ℓ, 0.02 pCi/ℓ and 0.02 pCi/ℓ, respectively. In November 1969, under higher discharge conditions, these levels were considerably lower.

Radionuclide concentrations in bed sediment were analyzed in May 1968, June 1969, and November 1969 (Table 8). The highest radionuclide concentrations except for ^{137}Cs and ^{239}Pu were found in Cattaraugus Creek at Springville Dam in the November 1969 survey. Concentrations of ^{106}Ru, ^{137}Cs, ^{134}Cs, ^{90}Sr, ^{238}Pu and ^{239}Pu were 190 pCi/g, 94 pCi/g, 29 pCi/g, 22 pCi/g, 2.3 pCi/g, 0.063 pCi/g, and 0.053 pCi/g, respectively. These values were generally higher than those found in Buttermilk Creek at Thomas Corners Bridge during the same sampling period.

TABLE 5. Radionuclide Concentrations in Cattaraugus and Buttermilk Creeks, June 1969 (U.S. Department of Health, Education and Welfare)

Location	Concentration in Water (pCi/ℓ)								
	H-3	Gross α	Gross β	Ru-106	Cs-137	Cs-134	Sr-90	Sb-125	Co-60
Buttermilk Creek, upstream	1,000	<1	110	110	<1	<0.5	<1	<2	<0.3
Cattaraugus Creek, upstream	1,000	<1	200	160	<1	<0.5	<1	<2	<0.3
Confluence Frank's and Buttermilk Creeks	280,000	3	5,900	4,000	54	18	420	35	4.3
Buttermilk Creek at Bond Road Bridge	400,000	4	7,400	5,400	30	13	480	41	5.3
Buttermilk Creek at Thomas Corners Bridge	370,000	4	6,800	4,700	16	5.2	480	41	4.9
Confluence of Buttermilk and Cattaraugus Creeks	340,000	4	6,200	4,300	16	4.7	440	40	4.4
Cattaraugus Creek at Felton Bridge	30,000	<1	560	430	1.3	<0.5	41	3.6	0.4
Cattaraugus Creek at Springville Dam	30,000	<1	530	410	1.2	<0.5	38	3.4	0.3
	Concentration in Suspended Solids (pCi/ℓ)								
Buttermilk Creek, upstream			13	10	1	0.5			
Cattaraugus Creek, upstream			14	11	1	0.5			
Confluence Frank's and Buttermilk Creeks			110	39	37	10	2		
Buttermilk Creek at Bond Road Bridge			120	36	38	11	3		
Buttermilk Creek at Thomas Corners Bridge			81	24	23	8.7	1		
Confluence of Buttermilk and Cattaraugus Creeks			200	120	23	7.9	1		
Cattaraugus Creek at Felton Bridge			67	45	1	0.5	1		
Cattaraugus Creek at Springville Dam			120	7	1	0.5	1		

TABLE 6. Radionuclide Concentrations in Cattaraugus and Buttermilk Creeks, November 1969 (U.S. Department of Health, Education and Welfare)

Location	Concentration in Water (pCi/ℓ)								
	H-3	Gross α	Gross β	Ru-106	Cs-137	Cs-134	Sr-90	Sb-125	Co-60
Buttermilk Creek, Thomas Corners Bridge	43,000	1.4	1,400	380	17	<10	360	49	<10
Cattaraugus Creek, Felton Bridge	3,400	<1	270	75	<10	<10	69	<40	<10
	Concentration in Suspended Solids (pCi/ℓ)								
Buttermilk Creek, Thomas Corners Bridge			320	64	142	33	<2		
Cattaraugus Creek, Felton Bridge			120	20	62	20	<1		

TABLE 7. Uranium and Plutonium Concentrations in Buttermilk Creek (U.S. Department of Health, Education and Welfare)

Date	Location	Concentration in Water (pCi/ℓ)				
		U-232	U-234	U-238	Pu-238	Pu-239
6/17/69	Buttermilk Creek at Thomas Corners Bridge	3.4	1.4	0.22	0.02	0.02
11/4/69	Buttermilk Creek at Thomas Corners Bridge	0.6	0.3	0.12	0.01	0.01
		Concentration Suspended Solids (pCi/ℓ)				
6/17/69	Buttermilk Creek at Thomas Corners Bridge	0.01	0.02	0.02	0.01	0.01
11/4/69	Buttermilk Creek at Thomas Corners Bridge				0.015	0.018

TABLE 8. Radionuclide Concentration in Bottom Sediments (U.S. Department of Health, Education and Welfare)

Date	Location	Concentration in Bed Sediment (pCi/gram)						
		Ru-106	Cs-137	Cs-134	Sr-90	Co-60	Pu-238	Pu-239
5/28/68	Buttermilk Creek at Thomas Corners Bridge	80	70	12	2.6	0.49	0.05	0.091
5/28/68	Cattaraugus Creek at Springville Dam	8	12	1.7	0.8	0.17	0.012	0.023
6/19/69	Buttermilk Creek at Thomas Corners Bridge	130	110	21	13	1.5	0.03	0.05
6/19/69	Cattaraugus Creek at Springville Dam	34	35	7.7	9.9	0.21	-	-
11/6/69	Buttermilk Creek at Thomas Corners Bridge	110	110	22	13	1.3	-	-
11/6/69	Cattaraugus Creek at Springville Dam	190	94	29	22	2.3	0.063	0.053

NUCLEAR FUELS SERVICES ENVIRONMENTAL MONITORING PROGRAM(6)

Nuclear Fuels Services (NFS) collects monthly composite water samples (Table 9) at Felton Bridge as part of an environmental monitoring program. The period of record shown in Table 9 is from January 1972 to June 1972 and from May 1974 to December 1976.

Radioactivity levels of gross α, gross β, ^{3}H, ^{89}Sr, ^{90}Sr, ^{106}Ru, ^{129}I, ^{134}Cs and ^{137}Cs in water have generally decreased since 1971. Table 9 indicates that there is a strong relationship between radioactivity in water and water discharge of Cattaraugus Creek. In other words, during periods of high-flow radionuclide concentrations in water tend to be low, whereas during low-flow conditions the radionuclide levels are higher.

NEW YORK STATE DEPARTMENT OF ENVIRONMENTAL CONSERVATION MONITORING(7)

The New York State Department of Environmental Conservation conducts an extensive environmental radioactivity monitoring program throughout the State of New York. Included in their monitoring program is the area surrounding the Nuclear Fuels Service Center at West Valley. Table 10 is a summary of results of quarterly analysis of radioactivity in 1971 and 1972.

SUMMARY OF PREVIOUS RADIOLOGICAL SURVEYS

Tables 11 and 12 summarize comparable data on radioactivity levels in water and bed sediment from previous radiological surveys. The two most striking observations of radioactivity in water are the dilution effect on radioactivity between Buttermilk Creek and Cattaraugus Creek, and except for the bed sediments in Springville Reservoir, the general decrease in radioactivity with time. Figure 4 is a comparison of NFS environmental monitoring data with water discharge of Buttermilk Creek for the same period. Although no distinction was made between dissolved radioactivity and suspended sediment radioactivity, there does appear to be an inverse relationship between radioactivity and discharge, especially for ^{90}Sr which is mostly transported in the dissolved phase.

TABLE 9. Monthly Composite Water Samples Cattaraugus Creek at Felton Bridge (Nuclear Fuels Services)

					pCi/ℓ				
	Gross α	Gross β	H-3	Sr-89	Sr-90	Ru-106	I-129	Cs-134	Cs-137
1971									
Jan	1.04	100	4,300		31.0				
Feb	1.16	93	19,800		31.5				
Mar	1.00	74	16,600		31.6				
Apr	1.70	108	3,600		22.8				
May	0.35	143	4,800		60.1				
Jun	0.84	108	8,100		23.7				
Jul	0.49	93	85,000		19.2		0.77		
Aug	1.73	194	84,700		32.6		1.34		
Sep	0.38	104	18,700		23.5		3.10		
Oct	0.75	184	34,500	0.00	37.3	55	0.65	0.00	2.8
Nov	0.84	183	49,300	0.00	25.6	152	0.54	0.00	5.9
Dec	3.05	143	3,700	0.00	17.2	75	0.03	0.00	15.6
1972									
Jan	0.59	61	3,500	0.00	11.1	138	0.02	0.00	27.5
Feb	0.51	130	3,900	0.00	29.3	193	0.03	0.00	66.1
Mar	3.05	140	1,600	0.00	23.3	253	0.02	0.00	34.1
Apr	0.93	47	29,100	0.00	19.5	379	0.88	0.00	53.5
May	0.91	114	4,600	0.00	14.2	183	0.20	0.00	25.0
Jun	3.23	40	3,600	0.00	11.8	128	0.01	0.00	7.0
1974									
May	<0.80	6.3	<1,900	0.00	2.4	23	0.00067	0.00	1.8
Jun	<1.08	6.7	<1,900	0.00	1.6	<14	---	0.00	4.0
Jul	1.34	11.6	<1,900	6.98	2.7	<14	---	0.00	0.0
Aug	<0.92	10.2	<1,900	0.00	5.0	<14	---	0.00	2.5
Sep	<1.12	21.0	<1,900	3.60	5.6	<14	0.00312	0.00	3.2
Oct	<0.94	13.9	<1,900	0.00	6.1	26	0.00911	0.00	0.0
Nov	<0.73	16.1	<1,900	0.00	2.9	36	0.00304	0.00	0.0
Dec	<1.17	5.3	<1,900	0.00	5.2	<14	---	0.00	0.0
1975									
Jan	<0.95	6.9	<1,900	0.00	0.0	<14	0.00363	0.00	0.0
Feb	<0.71	10.0	<1,900	0.00	0.0	<14	---	0.00	0.0
Mar	<0.30	<2.1	<1,900	0.00	0.0	<14	0.00491	0.00	0.0
Apr	<0.59	4.1	<1,900	0.00	1.2	<14	---	0.00	4.0
May	<0.40	5.3	<1,900	0.00	1.9	<14	---	0.00	4.5
Jun	<0.78	18.7	14,200	1.20	2.2	<14	0.00279	0.00	2.4
Jul	<0.60	5.2	1,900	5.80	1.8	<16	---	0.00	1.8
Aug	<0.60	13.5	3,900	0.00	5.4	20	---	0.00	2.1
Sep	<0.60	6.6	<700	0.00	1.7	15.9	---	0.00	0.0
Oct	<0.60	7.8	11,800	0.00	2.5	<16	---	0.00	0.0
Nov	<0.60	7.9	<700	0.00	3.4	<16	---	0.00	0.0
Dec	<0.60	4.4	19,500	0.00	0.0	37.8	---	0.00	0.0
1976									
Jan	---	---	---	---	---	---	---	---	---
Feb	<0.71	9.8	4,300	0.00	0.0	28.1	0.0067	0.00	0.0
Mar	0.63	3.7	<700	0.00	0.0	34.3	0.0036	0.00	0.0
Apr	<0.60	3.9	<700	0.00	0.0	21.6	0.0041	0.00	1.2
May	<0.60	<4.3	2,900	0.00	1.9	18.5	---	0.00	0.0
Jun	2.03	14.6	4,300	0.00	1.6	16.4	0.0206	0.00	0.0
Jul	4.25	12.9	4,800	0.00	2.8	15.4	---	0.00	0.0
Aug	2.96	16.0	29,000	1.1	0.0	15.1	0.0035	0.00	0.0
Sep	2.49	8.9	65,700	0.00	2.8	<14.2	0.0076	0.00	2.3
Oct	1.80	6.5	7,600	2.1	0.0	<13.4	<0.0005	0.00	0.0
Nov	1.36	9.6	5,300	0.00	4.2	14.7	0.0018	0.00	0.0
Dec	<0.81	6.4	70,100	3.6	1.7	---	0.0033	0.00	3.5

TABLE 10. Average Radioactivity Levels in Water - pCi/ℓ (New York State Department of Environmental Conservation)

	Cs-137	Ru-106	Sb-125	H-3	Gross β	Sr-89	Sr-90	Pu-238	Pu 239,240	I-129
1971 - 3rd Quarter										
Buttermilk Cr. at Thomas Corners Br.	60	3,977	576	1,000,000	5,167	-	797	0.026	0.068	4
Cattaraugus Cr. at Springville Dam	-	197	15	66,000	269	-	44			-
Cattaraugus Cr. at Gowanda	-	-	-	92,000	86	-				
Cattaraugus Cr. at Irving	-	-	-	47,000	115	-	18			
1971 - 4th Quarter										
Buttermilk Cr. at Thomas Corners Br.	-	2,756	416	330,000	3,299	9	243	0.047	0.014	2.8
Cattaraugus Cr. at Springville Dam	-	247	21	39,000	304	-	24			-
Cattaraugus Cr. at Gowanda										
Cattaraugus Cr. at Irving	-	-	-	20,000	106					
1972 - 1st Quarter										
Buttermilk Cr. at Thomas Corners Br.	-	773	151	25,000	1,434		123			-
Cattaraugus Cr. at Springville Dam	-	95	-	3,200	146		15			-
1972 - 2nd Quarter										
Buttermilk Cr. at Thomas Corners Br.	11	920	242	27,000	1,307		112	-	-	0.42
Cattaraugus Cr. at Springville Dam	-	83	25	2,000	117		8	-	-	0.30

TABLE 11. Summary of Previous Analyses of Radioactivity in Water (Reported as pCi/ℓ)

	Sr-90	Gross β	Cs-137	H-3	ZrNb 95	RuRh 106	Co-60	Gross α	Cs-134	Sb-125
Buttermilk Creek at Thomas Corners Bridge										
1965										
NYS Health Dept.	<3									
1966										
NYS Health Dept.	<3	5.8								
NYS Aquatic Survey			1		6					
1967										
NYS Health Dept.	335	970	112	320,430						
NYS Aquatic Survey			100		10	1000	10			
1969 June										
U.S. Public Health	480	6800	16	370,000		4700	4.9	4	5.2	41
1969 November										
U.S. Public Health	360	1400	17	43,000		380	<10	1.4	10	49
Cattaraugus Creek at Springville Dam										
1965										
NYS Health Dept.	3									
1966										
NYS Health Dept.	62	116								
NYS Aquatic Survey			1							
1967										
NYS Health Dept.	24	95	<20	30,730						
NYS Aquatic Survey			10			100	4			
1969 June										
U.S. Public Health	38	530	1.2	30,000		410	0.3	<1	<0.5	3.4

TABLE 12. Summary of Previous Analyses of Radioactivity in Bed Sediments (Reported as pCi/g)

Location	Sr-90	Cs-137	Ru-106	ZrNb 95	Co-60	Cs-134
BUTTERMILK CREEK AT THOMAS CORNERS BRIDGE						
1966						
NYS Health Dept.	81	126	318	155	7.7	
1967						
NYS Health Dept.	2.6					
NYS Aquatic Survey		115	226	120	8	
1968 May						
U.S. Public Health	2.6	70	80		0.49	12
1969 June						
U.S. Public Health	13	110	130		1.5	21
1969 November						
U.S. Public Health	13	110	110		1.3	22
CATTARAUGUS CREEK AT SPRINGVILLE DAM						
1966						
NYS Health Dept.	3.8	9.0	32	10.7	0.4	
1967						
NYS Health Dept.	2.3	23.0	2.9	3.9	0.3	
NYS Aquatic Survey		28.8	45.7	7.4	0.47	
1968 May						
U.S. Public Health	0.8	12	8		0.17	1.7
1969 June						
U.S. Public Health	9.9	35	34		0.21	7.7
1969 November						
U.S. Public Health	22	94	190		2.3	29

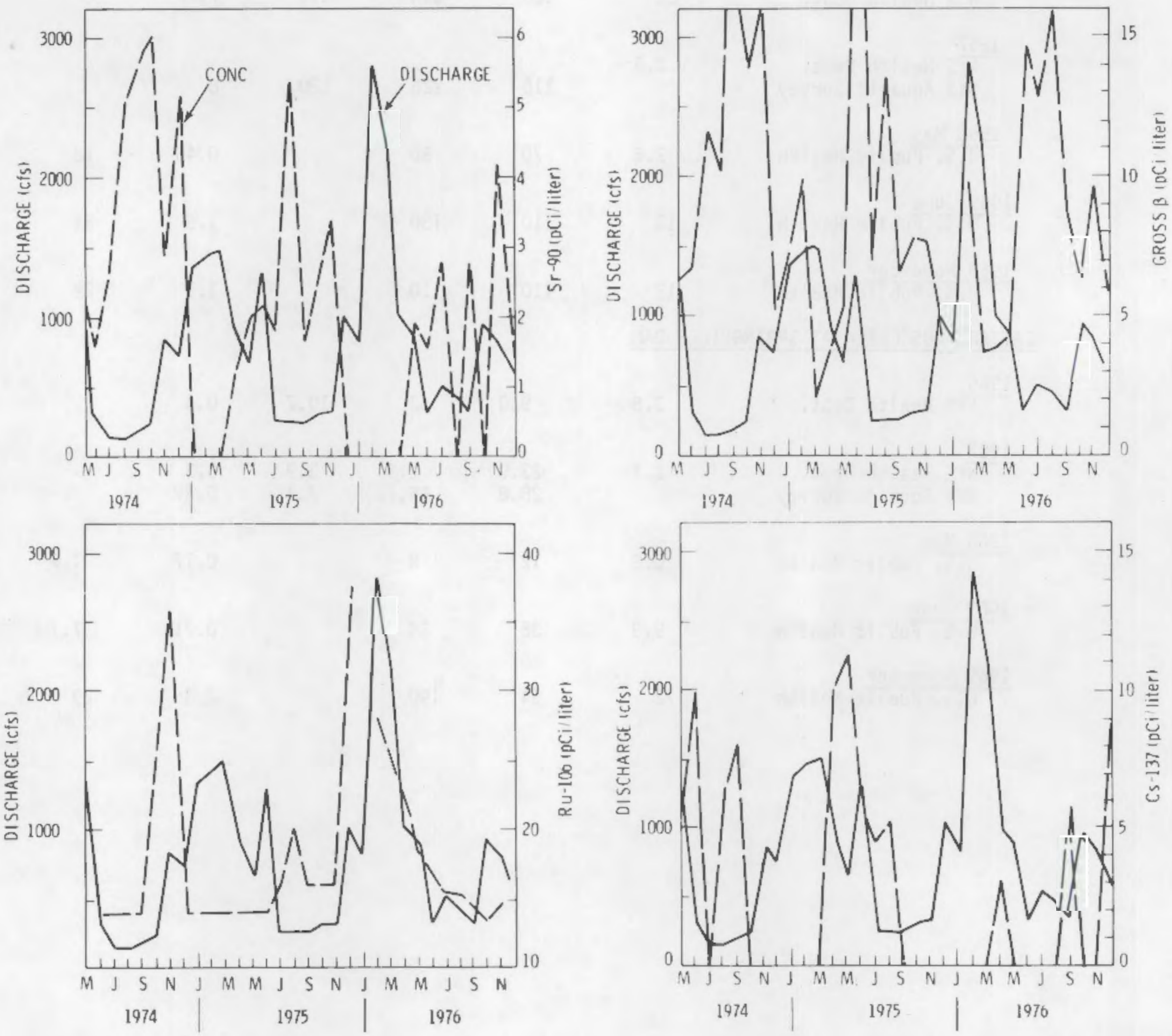

FIGURE 4. Summary - Comparison of NFS Environmental Monitoring Data with Water Discharge:
——— Discharge
- - - - - Radioactivity Levels

PHASE 1. SAMPLING PROGRAM

The Phase 1 sampling program was conducted on 30 November through 5 December 1977. The field sampling program consisted of collection of data on flow and channel characteristics, water characteristics, sediment characteristics, and radionuclide characteristics at designated transects on Cattaraugus and Buttermilk Creeks.

DESCRIPTION OF TRANSECTS

The field sampling effort involved the collection of data at ten transects on Cattaraugus and Buttermilk Creeks, covering approximately 45 linear miles of stream channel. The sampling effort was concentrated in a six-mile reach of Cattaraugus Creek and Buttermilk Creeks between Springville Reservoir on Cattaraugus Creek and the Nuclear Fuel Services site on Buttermilk Creek. However, two upstream control transects were established, one each on Cattaraugus and Buttermilk Creeks, and two downstream transects were located on Cattaraugus Creek at Gowanda and at the mouth of Cattaraugus Creek near Lake Erie. One transect was located on Franks Creek at its confluence with Buttermilk Creek, four transects were located on Buttermilk Creek, and five transects on Cattaraugus Creek. The location of these transects are shown on Figures 1 and 5 and are described in detail below:

FC-1 Confluence Franks and Buttermilk Creeks - This transect is located on Franks Creek about 100 ft upstream of the confluence with Buttermilk Creek at Buttermilk Creek mile (BCM) 2.27. The cross section was located on the downstream side of the Ohio Railroad culvert.

BC-1 Fox Valley Road - This site was used to provide background information on Buttermilk Creek. The cross section was located about 100 ft upstream of the Fox Valley Road culvert at BCM 4.98.

BC-2 Buttermilk Creek - This transect is located on Buttermilk Creek approximately 1,200 ft downstream from the confluence with Franks Creek at BCM 2.05.

BC-3 Bond Road Bridge - This transect is located on Buttermilk Creek about 100 ft down- stream of the abandoned Bond Road Bridge at BCM 1.34.

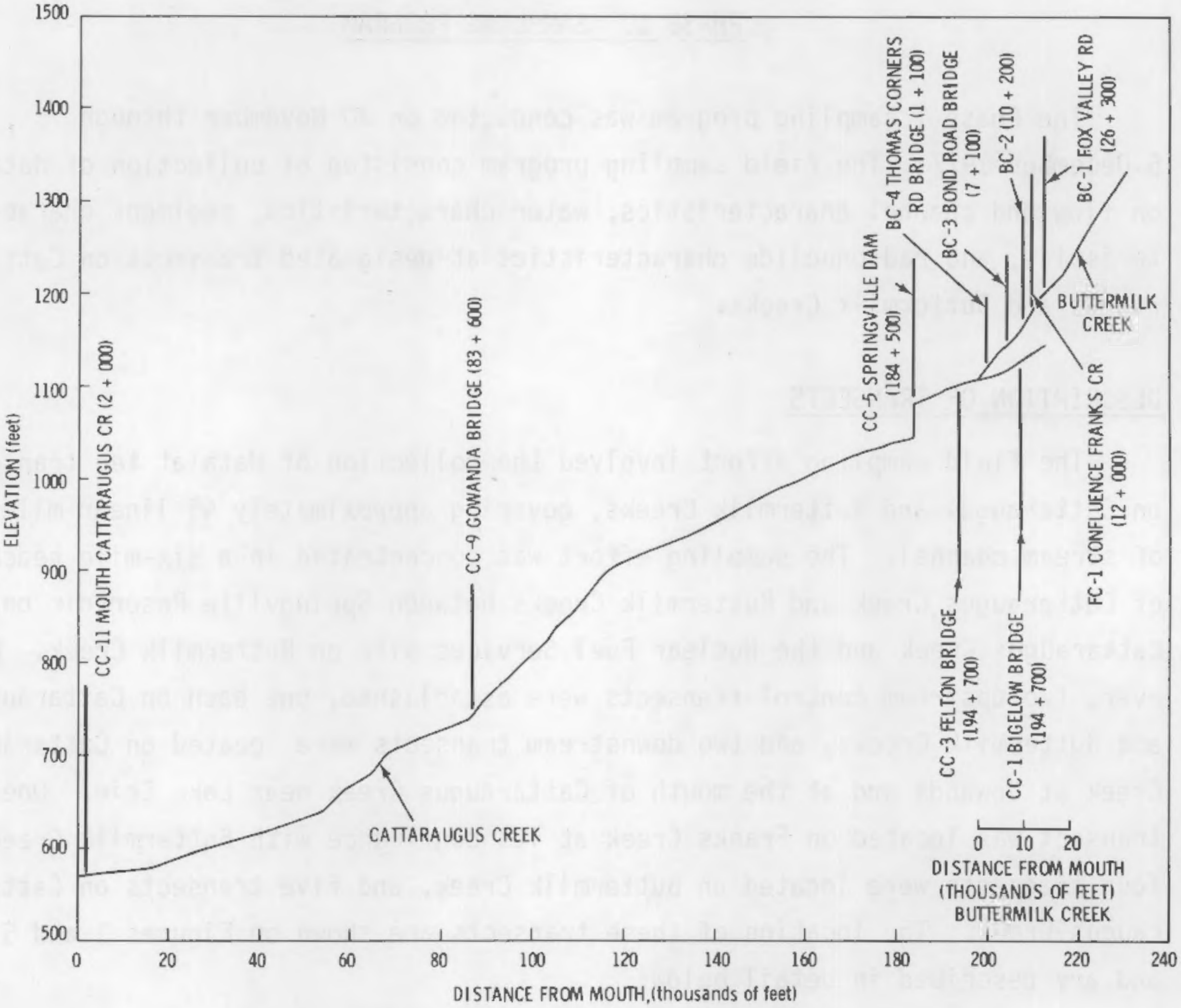

FIGURE 5. Location of Transects

BC-4 Thomas Corners Bridge - This transect is located on Buttermilk Creek about 100 ft downstream of Thomas Corners Bridge at BCM 0.21.

CC-1 Bigelow Bridge - This site, located on Cattaraugus Creek, was used to provide background information on Cattaraugus Creek. The cross section was located directly underneath the upstream side of the bridge at CCM 39.62.

CC-3 Felton Bridge - CC-3 is located on Cattaraugus Creek one-half mile downstream from the confluence with Buttermilk Creek. The cross section was located approximately 100 ft downstream of Felton Bridge at CCM 36.88.

CC-5 Springville Dam - This transect is located on Cattaraugus Creek in Springville Reservoir about 500 ft upstream of the dam. The cross section was located at CCM 35.04.

CC-9 Gowanda Bridge - This transect is located on Cattaraugus Creek at Gowanda. The cross section was located 150 ft downstream of the Taylor Hollow Road Bridge at CCM 15.83.

CC-11 Mouth Cattaraugus Creek - This transect is located on Cattaraugus Creek about 4000 ft upstream of the mouth. The cross section was situated underneath the New York-Central Railroad Bridge.

FLOW AND CHANNEL CHARACTERISTICS

Flow and channel characteristics data collected during the Phase 1 sampling program included cross section shape, channel bed and water surface elevations, and water velocity data at each transect. From these data the discharge could be calculated and bed degradation-aggradation between sampling periods determined. Cross-section shapes, and bed and water surface elevations were obtained using a level and surveying rod along pre-established transect lines. Bed elevations along the transect were taken so that the distance between any two measurements were less than 5% of the total channel width. Elevation measurements extended far enough beyond the confines of the existing stream channel to allow comparisons of the temporary reference datum points established near the transects. Water depths along some transects were too deep to use the survey level and rod technique to determine bed elevation. In such cases water depths were determined by means of a sounding rod or line and referenced to the water surface. The water surface elevations were referenced to the temporary reference datum point.

Water velocity measurements were obtained using electromagnetic induction current meters. Where the water depth was greater than 2.5 ft, velocity measurements were taken at two depths, 0.2 and 0.8 times the total water depth. Where the water depth was less than 2.5 ft, one velocity measurement was taken at 0.6 times the total water depth below the surface. From these velocity measurements the vertically averaged velocity can be calculated in order to determine the water discharge at the transect. Laterally across the stream, velocity measurements were taken at each location where the bed elevation was determined. The current meters were attched to wading rods where the water depths permitted; or where the depths were too great, a torpedo weight and cable were used.

WATER CHARACTERISTICS

Water characteristics measured during the sampling program were water temperature, pH and hardness. Water temperature is required to calculate viscosity for the SERATRA code. The pH and hardness are used in evaluating the adsorption and desorption coefficients.

Water temperature, pH, and hardness measurements were taken at predetermined stations and depths along the transects. Temperture measurements were taken using an in situ temperature probe and, pH and hardness samples were taken with a one-liter water sampler.

SEDIMENT CHARACTERISTICS

Total suspended sediment load samples were taken at designated stations and depths at each transect using a one-liter suspended sediment sampler.

RADIOLOGICAL SAMPLING

Large volume water samples, suspended sediment samples and bed sediment samples were collected at each transect for radiological analyses. The method for collection of the large volume water samples is shown in Figure 6. Approximately 80 gallons (300 liters) of water were collected in five gallon plastic containers using a centrifugal water pump, frozen and returned to the laboratory for separation of suspended solids from water. Standard filtering methods for separating suspended solids from water could not be used in the

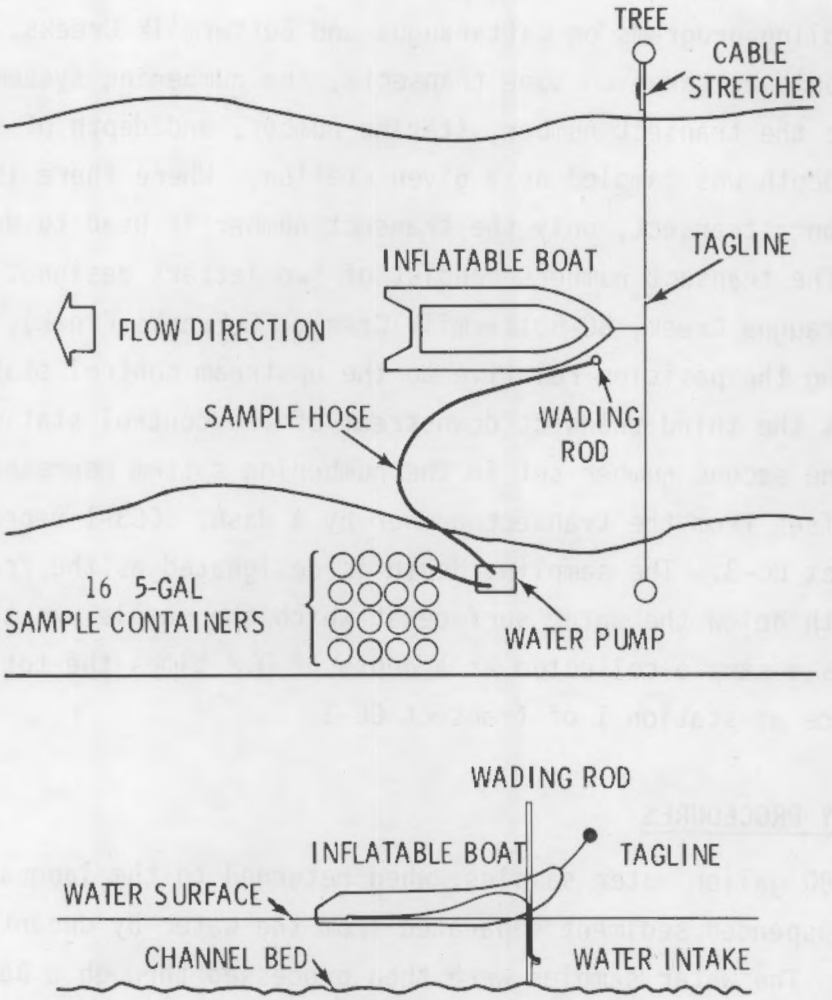

FIGURE 6. Sampling Procedure

field due to the high suspended sediment load (50 to >500 mg/ℓ) in Cattaraugus and Buttermilk Creeks, and because of the extreme difficulty in dividing suspended sediment into size fractions (i.e., sand, silt and clay) for radiological analysis once the suspended sediment is impregnated on filters.

SEDIMENT NUMBERING SCHEME

A uniform sample numbering system has been devised for use during the three sampling programs on Cattaraugus and Buttermilk Creeks. Since multiple samples could be taken on some transects, the numbering system has been set up to include the transect number, station number, and depth of sample where more than one depth was sampled at a given station. Where there is one sampling location on a transect, only the transect number is used to designate the sample. The transect numbers consist of two letters designating the stream (CC-Cattaraugus Creek, BC-Buttermilk Creek, FC-Franks Creek), and a number designating the position relative to the upstream control station. Thus, CC-3 represents the third transect downstream of the control station on Cattaraugus Creek. The second number set in the numbering system represents the station and is offset from the transect number by a dash. CC3-1 represents station 1 at transect CC-3. The sampling depth is designated as the fraction of the total depth below the water surface at which the sample was taken. CC3-1/.2d represents a sample collected at a depth of 0.2 times the total depth below the surface at station 1 of transect CC-3.

LABORATORY PROCEDURES

The 80 gallon water samples, when returned to the laboratory, were thawed and the suspended sediment separated from the water by decantation and centrifugation. The water samples were then processed through a Battelle Large Volume Water Sampler (BLVWS). The BLVWS is comprised of a set of three parallel pre-filters, three aluminum oxide beds and three cation exchange resin beds, all in series. The pre-filters (0.3μ glass-fiber) are designed to capture the colloidal material not previously collected by the decantation and centifugation process. After passing through the pre-filters, the water then passes through the three 11 in.-dia by 0.25 in.-deep, 100x200 mesh chromatographic grade, neutral aluminum oxide beds. Then the water passes through the three DOWEX 50Wx8, 20x50 mesh cation exchange resin beds of the same dimensions as the aluminum oxide beds. A flow meter was located at the discharge end of the BLVWS to record the volume of water passing through the BLVWS. The use of the BLVWS with aluminum oxide and cation exchange resin beds for collection of low level radionuclides has been discussed by Silker.(8)

The three aluminum oxide beds and three resin beds were stored in plastic bags and numbered in order of position in the series (e.g., 1, 2 or 3) for radiological analysis. The three pre-filters were set up to be analyzed as one sample since they were set up in parallel sets.

Preparation of suspended sediment samples for radiological analysis consisted of separation into sand, silt, and clay size fractions. The procedures used were those of Jackson.(9) The suspended sediment was first wet sieved through a U.S. Standard 200 mesh sieve. The material retained on the sieve was dried and weighed, and reported as sand (>74µ). The material passing through the sieve was dispersed using an electric mixer (ASTM Stirring Apparatus A)(10) and centrifuged at 750 rpm for 3.3 minutes. The material remaining in suspension after centrifugation was decanted, dried and weighed. This fraction was reported as clay (>4.0µ). The material remaining in the centrifuge cups after decantation of the suspended clay was dried, weighed, and reported as silt (74µ x 4.0µ).

Bed sediment samples were dried in an oven at 103°F and then sieved on a RoTap Shaker. The material retained on each screen was weighed and a portion saved for radiological analysis. The portion passing the 200 mesh screen was allowed to soak in distilled water overnight and separated into silt and clay fractions by the same method used for suspended sediment.

Total suspended sediment levels were measured from the one liter suspended sediment sample by filtration according to <u>Standard Methods</u>, No. 208D.(11) Gooch crucibles and Whatman GF/C filters were used to filter 100 milliliter samples which were dried for a minimum of two hours and weighed. The results are reported as milligrams per liter (mg/ℓ).

Water hardness and pH measurements were taken in the field. The pH measurements were taken using a Corning Model 3 portable pH meter. The meter was standardized with pH 7 buffer immediately prior to each measurement.

Water hardness was measured utilizing the procedures described in Standard Methods, No. 309B using commercial reagents. Results are reported as mg/ℓ of $CaCO_3$.

RADIOLOGICAL ANALYSIS

Radiological analyses were performed on sand, silt and clay size fractions of suspended sediment and bed sediment samples, and on filters, aluminum oxide and resin beds of water samples. Priority was given to gamma ray spectrometry analysis during the Phase 1 sampling program because of initial funding limitations and the relatively low costs of gamma ray spectrometry. Additional funding allowed the analysis of other radionuclides requiring radiochemical separation procedures, such as ^{90}Sr, ^{238}Pu, ^{239}Pu, ^{241}Am and ^{244}Cm. However, the high costs of radiochemical separation did not warrant the analysis of all samples for non-gamma radionuclides.

United States Testing, Inc., a commercial laboratory located in Richland, Washington, performed the majority of the gamma ray spectrometry, ^{90}Sr, ^{238}Pu and ^{239}Pu analyses of the Phase 1 samples. The University of Washington, Laboratory of Radiation Ecology also performed some gamma ray spectrometry, plutonium and strontium analysis, in addition to performing ^{241}Am, ^{244}Cm, and tritium analyses. PNL's Physical Sciences Department performed gamma ray spectrometry analysis of one sample (FC-1).

Gamma ray spectrometry analysis was performed using Ge(Li) diode systems, except the one sample analyzed by PNL's Physical Sciences Department which utilized a large-volume NaI(Tl) multi-dimensional gamma ray spectrometer system in the coincidence and noncoincidence modes.(12)

Gamma ray spectrometry analysis by U.S. Testing, Inc. utilized a Canberra Model 7229 and 7248, Germanium-Lithium drifted diode and a Pulse Height Analyses (PHA) program for intepretation of the Ge(Li) spectra. A description of the system is included in Appendix A of this report. The published detection limit of U.S. Testing's gamma ray spectrometry analysis is 0.07 pCi/g based on a 500 minute count and 40 grams of sample. For a 200 minute count the detection limit is 0.1 pCi/g. The above detection limits do not apply to all samples analyzed, however, since the sample sizes were often less than 40 grams. In many cases the suspended sand fraction and bed clay fraction quantities were less than one gram. In such cases the detection limits increased dramatically and can be seen in the results of the gamma ray spectrometry analyses where the standard error (represented as ± two standard deviations) is much greater than the detection limit.

A description of methods and procedures used by U.S. Testing, Inc., in their plutonium and strontium analysis of Phase 1 samples is also included in Appendix A.

Appendix B is a description of the analytical methods and procedures used by the University of Washington, Laboratory of Radiation Ecology in their gamma ray spectrometry, plutonium, strontium, americium and curium analyses of the Phase 1 samples.

DISCUSSION OF PHASE 1 SAMPLING PROGRAM RESULTS

Table 13 is a summary of the radiological samples collected during the 30 November through 5 December 1977 sampling program. Twenty one samples were collected at the ten transects on Cattaraugus, Buttermilk and Franks Creeks. Multiple station and depth samples were taken at some transects to investigate the vertical and lateral variability of radionuclide concentrations. The sample stations and depths for each transect are shown in Appendix C.

FLOW AND CHANNEL CHARACTERISTICS

A comparison of computed discharges from measured velocities and depths at each transect with the daily discharge of Cattaraugus Creek at the USGS Gowanda gaging station is given in Figure 7. During the sampling program of 30 November through 5 December 1977, the daily discharge of Cattaraugus Creek at Gowanda varied from a low of 800 cfs on 30 November to a high of 3700 cfs on 1 December. From 1 December to 5 December the discharge steadily decreased to 940 cfs. This peaking hydrograph, which occurred during the sampling program, was the result of snow melt and thawing conditions on 1 December, freezing conditions from 2 to 4 December, and heavy snowfall and freezing conditions on 5 December. The peaking hydrograph of Cattaraugus Creek is reflected in the discharge computations of the sampling transects.

High discharges were encountered at Buttermilk Creek transects, BC-2, BC-3, and BC-4, on 1 and 2 December because of the peaking hydrograph. These discharges were 360 cfs, 95 cfs and 139 cfs, respectively. The average discharge for Buttermilk Creek at Thomas Corners Bridge, based on eight years of record by the USGS is 46.5 cfs. The discharges of Franks Creek and Buttermilk Creek at Fox Valley Road during the Sampling Program were 20 cfs and 52 cfs, respectively.

Discharges of Cattaraugus Creek on 2 through 4 December at each of the sampling transects show the effects of the declining hydrograph. On 2 December the discharge of Cattaraugus Creek at Bigelow Bridge (CC-1), the upstream control transect on Cattaraugus Creek, was 845 cfs, whereas the discharge at CC-3, Felton Bridge, on 3 December was 722 cfs. The computed

TABLE 13. Summary of Work Conducted

Date	Transect		Number of Stations Sampled	Number of Depths Sampled	Total Number of Samples
11/30/77	FC-1	Franks Creek	1	1	1
11/30/77	BC-1	Fox Valley Road	1	1	1
12/1/77	BC-2	Buttermilk Creek	2	1,1	2
12/2/77	BC-3	Bond Road Bridge	2	1,1	2
12/2/77	BC-4	Thomas Corners Bridge	2	1,1	2
12/2/77	CC-1	Bigelow Bridge	1	1,1	1
12/3/77	CC-3	Felton Bridge	3	1,3,1	5
12/4/77	CC-11	Mouth Cattaraugus Creek	1	2	2
12/4/77	CC-9	Gowanda Bridge	1	1	1
12/5/77	CC-5	Springville Dam	2	1,3	4
Total			16		21

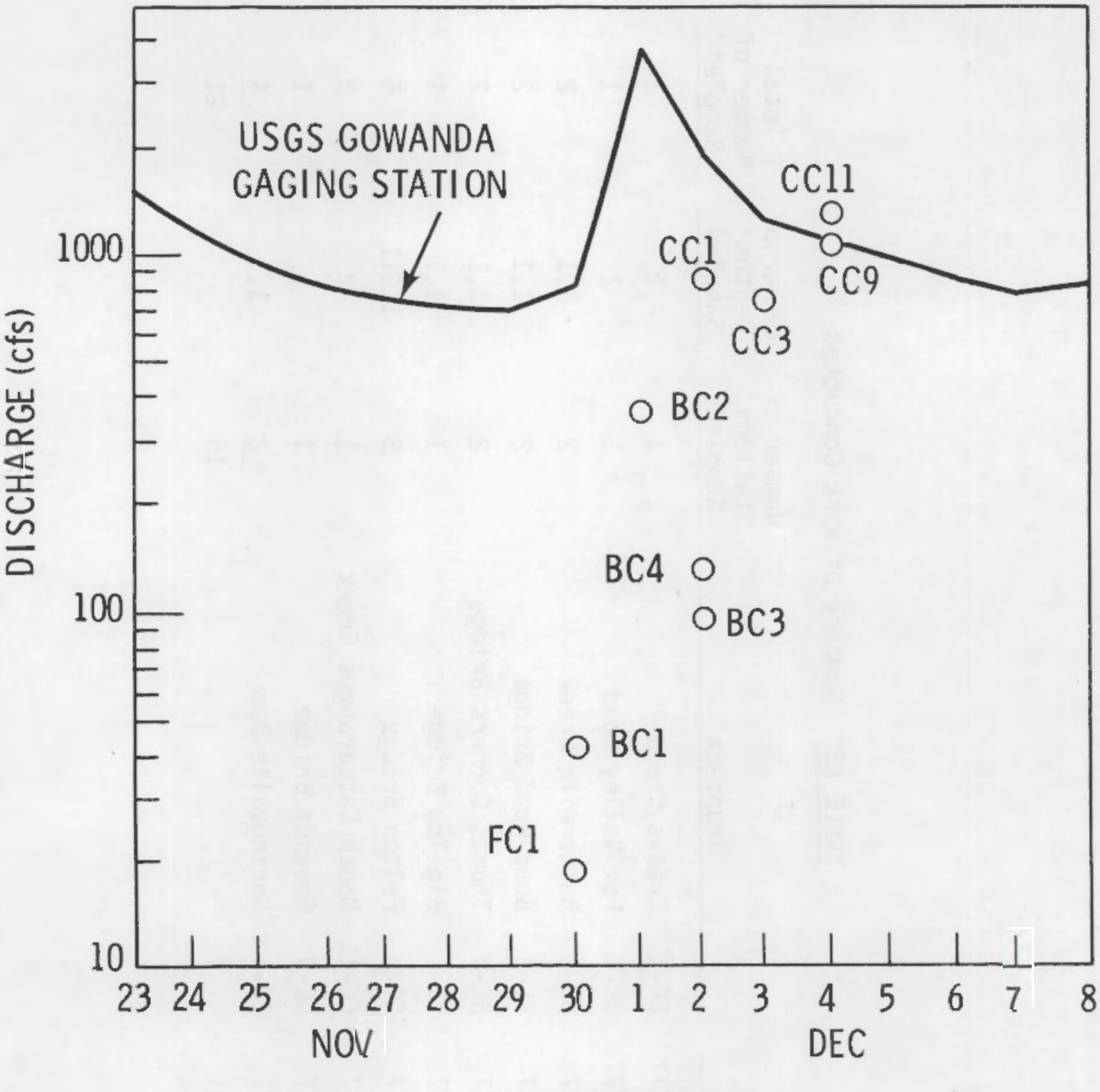

FIGURE 7. Daily Discharges of Cattaraugus Creek at USGS Gowanda Station and Sampling Transects

discharge at CC-9, Gowanda Bridge, located just downstream of the USGS gaging station, was 1100 cfs, the same as the recorded discharge at the Gowanda gaging station. The discharge at CC-11, mouth of Cattaraugus Creek, was 1326 cfs. The average discharge of Cattaraugus Creek at Gowanda, based on 37 years of record, is 731 cfs.

WATER CHARACTERISTICS

Water temperatures during the Phase 1 Sampling Program varied from a high of 5°C at BC-2 on 1 December to 1°C at CC-5, Springville Dam on 5 December (Table 14). No vertical temperature variations were discernible. Water hardness of samples varied from 29 to 68 mg/ℓ of $CaCO_3$. No distinguishable patterns were evident.

The pH of water varied from 6.9 to 9.2. The highest values were found in Franks Creek and Buttermilk Creek (7.5 to 9.2) and the lowest in Cattaraugus Creek (6.9 to 8.0).

SEDIMENT CHARACTERISTICS

Suspended sediment loads varied from about 1400 mg/ℓ in Franks Creek to 45 mg/ℓ in Buttermilk Creek at Thomas Corners Bridge (as shown in Table 15). Suspended sediment load of Buttermilk Creek was generally higher than in Cattaraugus Creek and can be partially accounted for by the higher discharges during the period of sample collection at these transects.

Suspended sediment was comprised principally of silt and clay with very little sand. In most cases the suspended sand accounted for less than 10% of the suspended sediment load. Suspended silt generally accounted for greater than 70% of the suspended sediment load and suspended clay accounted for from 5% to 10% of the suspended sediment load.

In contrast, the bed sediment (Table 16) was comprised almost exclusively of sand and gravel size material with very little silt and almost no clay size sediment.

TABLE 14. Water Characteristics and Discharge

Transect/Station/Depth	Date Sampled	Depth (ft)	Average Velocity fps	Computed Discharge cfs	Water Temperature °C	Hardness mg/ℓ $CaCO_3$	pH
FC-1 Franks Creek	11/30/77	1.33	1.25	20			
0.2 d		0.54			3.0	46	8.5
BC-1 Fox Valley Road	11/30/77	0.76	2.63	52			
0.2 d		0.54			3.0	54	8.9
BC-2 Buttermilk Creek	12/01/77		3.73	360			
Station 1		2.15					
0.2 d		1.52			5.0	41	8.5
Station 2		0.79					
0.2 d		0.54			5.0	43	7.5
BC-3 Bond Road Bridge	12/02/77		2.02	95			
Station 1		0.86					
0.2 d		0.65			4.0	48	8.6
Station 2		1.11					
0.2 d		0.82			4.0	29	9.2
BC-4 Thomas Corners Bridge	12/02/77		2.31	139			
Station 1		0.46					
0.2 d		0.32			4.0	56	8.8
Station 2		1.73					
0.2 d		1.56			4.0	29	8.3
CC-1 Bigelow Bridge	12/02/77		3.09	845			
Station 1		2.23					
0.2 d		1.73			4.0	44	7.1
CC-3 Felton Bridge	12/03/77		3.22	722			
Station 1		3.25					
0.2 d		0.67			4.0	42	7.4
0.5 d		1.58			4.0	42	7.0
0.8 d		2.58			4.0	50	8.0
Station 2		2.17					
0.2 d		1.42			4.0	34	7.0
Station 3		1.71					
0.2 d		0.96			4.0	46	7.5
CC-5 Springville Dam	12/05/77						
Station 1		11.5					
0.2 d		9.2			1.5	54	7.2
Station 2		11.5					
0.2 d		2.3			1.0	47	6.9
0.5 d		5.8			1.0	50	7.1
0.8 d		9.2			1.0	54	6.9
CC-9 Gowanda Bridge	12/04/77		2.86	1100			
Station 1		4.2					
0.2 d		3.4			3.0	68	7.0
CC-11 Mouth Cattaraugus Cr.	12/04/77		0.67	1326			
Station 1		7.0					
0.2 d		1.4			3.5	54	7.0
0.8 d		5.6			3.5	52	7.1

0.2 d
0.5 d } indicate fraction of total depth below surface, i.e., 0.8 d indicates 0.8 x the water depth below the surface
0.8 d

TABLE 15. Suspended Sediment Characteristics

		% Sand	% Silt	% Clay	Total Suspended Sediment load, mg/ℓ
FC-1	Franks Creek	0.6	88.5	10.9	1392
BC-1	Fox Valley Road	2.0	89.1	8.9	360
BC-2	Buttermilk Creek				
	Station 1	18.4	78.1	3.4	570
	Station 2	9.1	62.9	28.1	570
BC-3	Bond Road Bridge				
	Station 1	2.5	88.7	8.8	125
	Station 2	19.0	74.7	6.3	54
BC-4	Thomas Corner Bridge				
	Station 1	3.9	86.4	9.7	50
	Station 2	1.5	86.7	11.8	45
CC-1	Bigelow Bridge	4.6	90.7	4.7	55
CC-3	Felton Bridge				
	Station 1				
	0.2d	7.0	89.1	3.9	74
	0.5d	9.4	85.9	4.7	60
	0.8d	9.2	87.0	3.8	70
	Station 2	0.9	47.4	51.7	81
	Station 3	1.1	47.0	51.9	84
CC-5	Springville Dam				
	Station 1	0.8	90.2	9.0	79
	Station 2				
	0.2d	0.6	91.0	8.4	97
	0.5d	0.6	99.4	0.0	105
	0.8d	0.6	89.5	9.9	104
CC-9	Gowanda Bridge	10.0	84.3	5.7	72
CC-11	Mouth Cattaraugus Cr.				
	0.2d	2.6	93.2	1.2	85
	0.8d	9.8	85.2	5.0	73

TABLE 16. Bed Sediment Characteristics

		% Sand	% Silt	% Clay
FC-1	Franks Creek	28.1	68.5	3.4
BC-1	Fox Valley Road	98.5	1.3	0.2
BC-2	Buttermilk Creek			
	Station 1	98.9	1.1	0.01
	Station 2	99.2	0.7	0.003
BC-3	Bond Road Bridge			
	Station 1	99.4	0.5	0.01
	Station 2	98.2	1.8	0.01
BC-4	Thomas Corners Bridge			
	Station 1	99.9	0.1	---
	Station 2	99.1	0.9	0.03
CC-1	Bigelow Bridge	99.8	0.2	0.02
CC-3	Felton Bridge			
	Station 1	99.5	0.5	0.003
	Station 2	98.7	1.3	0.004
	Station 3	99.8	0.2	0.003
CC-5	Springville Dam			
	Station 1	89.0	10.7	0.3
	Station 2	81.0	18.7	0.3
CC-9	Gowanda Bridge	98.9	1.1	0.1
CC-11	Mouth Cattaraugus Creek	87.4	11.3	1.3

GAMMA RAY SPECTROMETRY

Tables 17 through 21 are the results of gamma ray spectrometry analysis of radionuclides attached to suspended sediment, bed sediment, and radionuclides dissolved in water. The gamma ray spectrometry results of suspended and bed sediment are reported as the activity of a particular radionuclide associated with the sand, silt and clay fractions of each sample in picocuries per gram (pCi/g). To arrive at a composite sample activity for a particular radionuclide, the weight or percent of each size fraction is multiplied by the activity of that fraction, summed and divided by the total sample weight (divide by 100 in the case of percent). Gamma ray spectrometry results of radionuclides dissolved in water are reported as the total activity of a particular radionuclide (Table 19) and as the activity associated with each of the three resin beds, aluminum oxide beds and filters (Table 20).

Transects BC-1 and CC-1 are the upstream control stations on Buttermilk and Cattaraugus Creeks. Because these transects are upstream of the influence of the Nuclear Fuel Services, Inc. (NFS) complex at West Valley, New York, the radioactivity associated with the surface waters at these transects can be considered to be background. By comparing the gamma activity of suspended sediment, bed sediment and water at the upstream control transects with the activity downstream of the NFS complex, one can, with a certain degree of confidence determine the influence of NFS facility on the radioactivity in Cattaraugus and Buttermilk Creeks.

The principal gamma radionuclides detected by U.S. Testing, Inc., in the Phase 1 samples were ^{137}Cs, ^{134}Cs, ^{60}Co, ^{95}ZrNb, ^{155}Eu, ^{144}Ce, ^{141}Ce, ^{224}Ra, ^{226}Ra, ^{106}Ru, and ^{40}K. In addition, the University of Washington found detectable quantities of ^{101}Rh, ^{125}Sb, ^{232}Th, ^{228}Th, ^{235}U, ^{238}U, and ^{210}Pb in some samples.

Gamma Activity Associated with Suspended Sediment

Table 17 and portions of Table 21 summarize the results of gamma ray spectrometry analysis of suspended sediment. The majority of the suspended sediment load was comprised of the silt size fraction. This resulted in a variable geometry when counting by gamma ray spectrometry. As a consequence

TABLE 17. Gamma Ray Spectrometry - Radionuclides Attached to Suspended Sediment (pCi/g)

Transect/ Station/ Depth	Fraction	Weight of Sample, gm	Cs-137	Cs-134	Co-60	ZrNb-95	Eu-155	Ce-144	Ce-141	Ra-224	Ra-226	Ru-106	K-40
FC-1													
	Sand	0.9	10.5(2.1)(1)	0.24(0.19)(2)	<0.100(2)	N.D.(1)	N.A.	N.D.(1)	N.A.	N.A.	N.D.(1)	N.D.(1)	N.D.
	Silt	132.5	5.17(0.26)(1)	0.12(0.02)(2)	0.05(0.1)(2)	N.D.(1)	N.A.	N.D.(1)	N.A.	N.A.	1.04(0.11)(1)	N.D.(1)	16.9(2.8)(1)
	Clay	16.4	11.3(0.6)(1)	0.17(0.03)(2)	0.07(0.01)(2)	N.D.(1)	N.A.	N.D.(1)	N.A.	N.A.	1.17(0.24)(1)	N.D.(1)	42(6.6)(1)
BC-1													
	Sand	2.2	0.63(2)	<0.06(2)	<0.038(2)	N.D.(1)	N.A.	N.D.(1)	N.A.	N.A.	N.D.(1)	N.D.(1)	37.5(31.9)(1)
	Silt	96.8	0.11(0.03)(2)	<0.016(2)	<0.005(2)	N.D.(1)	N.A.	0.70(0.52)(1)	N.A.	N.A.	0.99(0.15)(1)	N.D.(1)	19.1(4.2)(1)
	Clay	9.7	0.36(0.11)(2)	<0.030(2)	<0.001(2)	N.D.(1)	N.A.	N.D.(1)	N.A.	N.A.	1.32(0.35)(1)		39.5(9.6)(1)
BC2-1													
	Sand	20.1	7.64(0.47)	<0.171	<1.44	<0.026	N.D.	N.D.	N.D.	<0.241	<0.576	<0.172	9.10(5.8)
	Silt	85.1	0.841(0.09)	<0.030	<0.004	N.D.	N.D.	<0.210	N.D.	<0.928	<0.692	<0.118	14.2(1.7)
	Clay	3.8	3.10(1.5)	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	<1.34	<3.63	33.1(31)
BC2-2													
	Sand	13.7	14.50(0.82)	<0.344	<0.259	1.30(0.71)	57.5(1.8)	N.D.	N.D.	N.D.	<1.27	3.32(3.2)	10.1(9.5)
	Silt	95.6	1.50(0.10)	<0.026	N.D.	N.D.	N.D.	<0.148	<0.029	0.844(0.13)	0.546(0.13)	N.D.	11.7(1.5)
	Clay	42.8	1.65(0.19)	<0.043	N.D.	<0.191	3.53(0.45)	N.D.	N.D.	N.D.	0.809(0.31)	<0.556	11.6(3.3)
BC3-1													
	Sand	0.5	<7.24	0.265	<0.332	<0.647	N.D.	N.D.	N.D.	N.D.	<0.467	<1.94	4.19
	Silt	18.1	2.55(0.38)	0.09	<0.016	N.D.	N.D.	N.D.	N.D.	1.13(0.64)	1.12(0.63)	<0.470	22.1(6.8)
	Clay	1.8	7.80(3.7)	0.772	N.D.	<1.49	N.D.	N.D.	N.D.	N.D.	<1.32	<6.13	<17.2
BC3-2													
	Sand	8.1	13.6(1.1)	<0.391	<0.105	N.D.	N.D.	N.D.	N.D.	N.D.	<0.562	<1.10	<11.4
	Silt	31.9	3.26(0.29)	<0.116	<0.072	<0.297	5.92(0.61)	N.D.	N.D.	N.D.	0.982(0.40)	N.D.	20.3(4.5)
	Clay	2.7	7.37(2.4)	<0.789	<0.644	<0.619	N.D.	N.D.	N.D.	N.D.	<1.13	<1.01	<3.08
BC4-1													
	Sand	0.9	<4.50	<1.16	<0.144	<0.0431	N.D.	N.D.	N.D.	N.D.	<0.015	N.D.	<1.99
	Silt	10.8	2.67(0.37)	<0.024	N.D.	N.D.	N.D.	<0.164	N.D.	0.697(0.61)	0.789(0.59)	N.D.	20.5(6.5)
	Clay	2.1	6.55(3.0)	<1.77	<1.36	<4.48	73.4(8.8)	23.3(15)	5.75(4.3)	5.06	<6.73(5.7)	36.6(20)	97.5(61)
BC4-2													
	Sand	0.3	0.316	0.425	<0.646	N.D.	N.D.	N.D.	N.D.	N.D.	<9.08	<1.02	<8.35
	Silt	16.9	2.41(0.44)	0.075	N.D.	<0.286	3.17(1.1)	N.D.	N.D.	N.D.	1.04(0.74)	N.D.	26.9(8.2)
	Clay	2.3	4.48(2.5)	0.115	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	<1.82	N.D.	<41.5
CC1													
	Sand	1.8	<1.38	<0.086	<0.251	<0.104	N.D.	N.D.	N.D.	N.D.	<0.141	<0.465	<1.52
	Silt	36.0	0.338(0.26)	<0.113	N.D.	<0.035	N.D.	N.D.	N.D.	<0.174	0.752(0.52)	<0.520	12.7(5.6)
	Clay	1.85	<0.264	N.D.	<0.421	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	<0.232	<30.8

(1) University of Washington
(2) PNL
Remaining samples analyzed by U.S. Testing
N.D. - Not detected
N.A. - Not analyzed
( ) - Two standard deviations

TABLE 17. (contd)

Transect/ Station/ Depth	Fraction	Weight of Sample, gm	Cs-137	Cs-134	Co-60	ZrNb 95	Eu-155	Ce-144	Ce-141	Ra-224	Ra-226	Ru-106	K-40
CC3-1/.2d													
	Sand	2.2	<1.09	<0.107	<0.430	N.D.	<1.07	N.D.	N.D.	N.D.	N.D.	N.D.	<11.2
	Silt	27.3	0.389(0.21)	<0.024	N.D.	N.D.	N.D.	<0.104	N.D.	<0.380	0.771(0.42)	0.107	11.2(4.4)
	Clay	1.2	<3.97	<3.40	<3.17	<6.59	<9.04	<52.9	<15.0	<16.4	<9.92	<73.8	156(110)
CC3-1/.5d													
	Sand	2.6	<1.07	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	<0.932	N.D.	<12.0
	Silt	23.9	0.400(0.27)	<0.077	<0.036	<0.081	<2.22(0.76)	<0.044	<0.098	N.D.	0.765(0.53)	<0.625	12.4(5.7)
	Clay	1.3	<1.04	<0.008	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	<67.4
CC3-1/.8d													
	Sand	2.8	9.02(2.2)	<0.029	<0.038	N.D.	N.D.	N.D.	N.D.	N.D.	<0.992	<0.091	<0.501
	Silt	26.5	0.477(0.22)	N.D.	<0.012	<0.071	N.D.	N.D.	N.D.	0.440(0.43)	0.564(0.42)	<0.105	12.4(4.6)
	Clay	1.2	NA	NA	NA	NA	NA	NA	NA				
CC3-2													
	Sand	0.5	<5.41	<0.104	<0.074	<0.017	N.D.	N.D.	N.D.	N.D.	<0.099	N.D.	<0.281
	Silt	26.9	0.385(.21)	N.D.	N.D.	<0.010	<0.046	N.D.	N.D.	<0.138	0.712(0.41)	N.D.	10.6(4.4)
	Clay	29.3	0.198(.19)	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	<0.231	N.D.	<2.80
CC3-3													
	Sand	0.6	<4.66	<1.02	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	<2.47	N.D.	<36.2
	Silt	25.3	0.326(0.22)	<0.090	N.D.	N.D.	N.D.	N.D.	N.D.	0.163	0.670(4.4)	N.D.	9.20(4.7)
	Clay	27.9	0.253(0.22)	<0.070	<0.032	<0.049	<0.383	N.D.	N.D.	N.D.	N.D.	N.D.	<4.54
CC5-1													
	Sand	0.2	<10.5	<0.462	<0.664	<0.260	<26.6	N.D.	N.D.	N.D.	N.D.	N.D.	<38.2
	Silt	23.8	0.887(0.29)	<0.006	<0.067	<0.104	2.26(0.76)	<0.244	N.D.	N.D.	0.591(0.53)	N.D.	7.86(5.5)
	Clay	2.4	5.46(2.7)	<0.233	N.D.	<0.611	17.5(7.5)	N.D.	N.D.	N.D.	<0.459	<0.892	<23.9
CC5-2/.2d													
	Sand	0.2	<0.320	<6.72	N.D.	N.D.	N.D.	N.A.	N.A.	N.D.	N.D.	<201.0	312.0
	Silt	29.3	0.630(0.21)	N.D.	N.D.	<0.017	N.D.	N.A.	N.A.	0.428(0.39)	0.616(0.38)	<0.189	12.1(4.1)
	Clay	2.7	3.35(2.1)	N.D.	N.D.	N.D.	N.D.	N.A.	N.A.	N.D.	<1.13	N.D.	<279.0
CC5-2/.5d													
	Sand	0.2	<0.412	<10.4	<1.36	<2.69	<10.5	N.D.	N.D.	N.D.	N.D.	N.D.	<38.2
	Silt	32.1	0.690(.39)	<0.025	N.D.	<0.086	<0.260	N.D.	N.D.	N.D.	0.650(0.39)	<0.309	11.0(4.2)
	Clay	--	NA	NA	NA	NA	NA	NA	NA				
CC5-2/.8d													
	Sand	0.2	NA	NA	NA	NA	NA	NA	NA				
	Silt	28.2	0.901(0.25)	<0.087	N.D.	<0.180	3.76(0.66)	N.D.	N.D.	N.D.	0.475(0.44)	<0.182	11.0(4.8)
	Clay	3.1	3.70(2.0)	<0.507	<0.183	N.D.	<1.07	N.D.	N.D.	N.D.	<0.873	<2.04	<35.0
CC9-1													
	Sand	2.6	<0.860	<0.006	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	<0.001	N.D.	<12.5
	Silt	22.0	0.290(0.29)	<0.061	<0.012	<0.076	N.D.	N.D.	N.D.	N.D.	<0.519	<1.03	11.4(6.1)
	Clay	1.5	<1.63	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	<0.649	<0.220	<34.1
CC11-1/.2d													
	Sand	0.9	<1.35	<0.080	<0.067	N.D.	N.D.	N.A.	N.A.	N.D.	<0.814	N.D.	<21.5
	Silt	30.8	0.454(0.23)	<0.018	N.D.	N.D.	N.D.	N.A.	N.A.	0.328	0.515(0.44)	<0.034	12.8(4.8)
	Clay	0.5	<2.60	<0.973	N.D.	N.D.	N.D.	N.A.	N.A.	N.D.	<1.44	N.D.	<1.52
CC11-1/.8d													
	Sand	--	NA	NA	NA	NA	NA	NA	NA				
	Silt	23.0	0.571(0.27)	<0.105	<0.013	<0.075	N.D.	N.D.	N.D.	N.D.	0.664(0.52)	<0.830	10.8(5.5)
	Clay	1.2	5.50(4.7)	<0.026	<0.007	N.D.	N.D.	N.D.	N.D.	N.D.	<1.65	N.D.	<15.2

N.D. - Not detected
N.A. - Not analyzed
( ) - Two standard deviations

TABLE 18. Gamma Ray Spectrometry - Radionuclides Attached to Bed Sediment (pCi/g)

		% of Total Sample by Weight	Cs-137	Cs-134	Co-60	ZrNb 95	Eu-155	Ce-144	Ce-141	Ra-224	Ra-226	Ru-106	K-40
FC-1													
	Sand	28.1	1.04(0.11)$^{(1)}$	0.16(0.03)$^{(2)}$	0.154(0.02)$^{(2)}$	N.D.$^{(1)}$	N.A.	N.D.$^{(1)}$	N.A.	N.A.	0.51(0.08)$^{(1)}$	N.D.$^{(1)}$	9.4(1.9)$^{(1)}$
	Silt	68.5	0.37(0.09)$^{(1)}$	<.006$^{(2)}$	<0.004$^{(2)}$	N.D.$^{(2)}$	N.A.	N.D.$^{(1)}$	N.A.	N.A.	0.87(0.10)$^{(1)}$	N.D.$^{(1)}$	15.0(2.7)$^{(1)}$
	Clay	3.4	1.11(0.11)$^{(1)}$	<.008$^{(2)}$	0.02(0.004)$^{(2)}$	0.47(0.39)$^{(1)}$	N.A.	0.58(0.32)$^{(1)}$	N.A.	N.A.	1.53(0.11)$^{(1)}$	1.33(0.69)$^{(1)}$	34.6(3.1)$^{(1)}$
BC-1													
	Sand	98.5	<0.005$^{(2)}$	<.002$^{(2)}$	<0.0004$^{(2)}$	N.D.$^{(1)}$	N.A.	N.D.$^{(1)}$	N.A.	N.A.	0.61(0.07)$^{(1)}$	N.D.$^{(1)}$	9.6(1.0)$^{(1)}$
	Silt	1.3	0.07(0.03)$^{(2)}$	<0.17$^{(2)}$	<0.005$^{(2)}$	N.D.$^{(1)}$	N.A.	N.D.$^{(1)}$	N.A.	N.A.	1.33(0.17)$^{(1)}$	2.02(1.23)$^{(1)}$	19.7(4.2)$^{(1)}$
	Clay	0.2	0.16(0.08)$^{(2)}$	<0.28$^{(2)}$	<0.001$^{(2)}$	N.D.$^{(1)}$	N.A.	N.D.$^{(1)}$	N.A.	N.A.	1.61(0.24)$^{(1)}$	2.3(1.8)$^{(1)}$	32.6(6.5)$^{(1)}$
BC2-1													
	Sand	98.9	3.74(0.16)	<0.105(0.085)	<0.050	N.D.	N.D.	<0.172	<0.085	0.923(0.17)	0.675(0.17)	N.D.	7.63(1.8)
	Silt	1.1	7.84(0.41)	<.174	<0.222	0.648(0.43)	26.1(0.88)	N.D.	N.D.	N.D.	1.47(0.46)	1.90(1.6)	10.2(4.7)
	Clay	0.01	73.5(21)	<0.104	<1.8	N.D.	N.D.	N.D.	N.D.	N.D.	<13.8	N.D.	<4.0
BC2-2													
	Sand	99.2	5.81(0.26)	0.143(0.13)	N.D.	0.521(0.24)	19.8(0.54)	N.D.	N.D.	N.D.	0.709(0.26)	1.52(0.90)	7.42(2.7)
	Silt	0.7	13.6(0.58)	<0.186	<0.140	<0.101	N.D.	N.D.	N.D.	<0.441	0.900(0.57)	<0.505	7.91(5.8)
	Clay	0.003	59.1(29)	<0.847	<5.99	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	<8.56
BC3-1													
	Sand	99.4	11.9(0.42)	<0.181	<0.124	N.D.	N.D.	N.D.	N.D.	0.996(0.37)	0.875(0.36)	<1.29	7.91(3.7)
	Silt	0.5	25.8(0.81)	0.362(0.33)	<0.308	<0.075	N.D.	N.D.	N.D.	0.725(0.63)	1.15(0.63)	N.D.	11.3(6.5)
	Clay	0.004	125(31)	<0.605	<1.50	<0.927	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	<9.0
BC3-2													
	Sand	98.2	7.04(0.20)	0.091(0.07)	<0.070	N.D.	N.D.	<0.122	<0.026	0.560(0.15)	0.482(0.15)	N.D.	7.01(1.6)
	Silt	1.8	10.9(0.31)	0.174(0.12)	<0.067	N.D.	N.D.	<0.098	N.D.	0.777(0.24)	0.756(0.24)	<0.474	9.66(2.5)
	Clay	0.01	40.7(8.1)	<0.695	<0.342	<5.91	N.D.	N.D.	N.D.	N.D.	<0.893	<8.01	<4.01
BC4-1													
	Sand	99.9	125(7.4)	<2.83	<1.19	11.9(7.9)	482(17)	N.D.	N.D.	N.D.	16.8(8.6)	33.2(30)	161(89)
	Silt	0.1	10.1(.48)	0.254(.25)	<.114	1.05(.47)	41.0(1.1)	N.D.	N.D.	N.D.	0.74(.50)	2.27(1.8)	13.2(5.3)
	Clay	--											
BC4-2													
	Sand	99.1	0.463(0.22)	<0.016	N.D.	N.D.	N.D.	N.D.	<0.016	N.D.	<0.142	N.D.	<466
	Silt	0.9	26.9(1.2)	<0.504	<0.345	N.D.	N.D.	N.D.	N.D.	<0.027	1.76(1.2)	<0.619	16.4(12)
	Clay	0.03	92.4(24)	<3.13	<2.58	<8.59	N.D.	N.D.	N.D.	N.D.	<2.12	<18.9	<10.6
CC-1													
	Sand	98.8	<0.042	<0.043	N.D.	N.D.	N.D.	<0.088	<0.002	0.431(0.12)	0.537(0.12)	N.D.	4.96(1.2)
	Silt	0.2	<0.087	<0.004	<0.009	N.D.	N.D.	N.D.	N.D.	0.743(0.32)	0.953(0.33)	<0.067	9.14(3.4)
	Clay	0.02											

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(1) University of Washington
(2) PNL
Remaining Samples analyzed by U.S. Testing
N.D. - Not detected
N.A. - Not analyzed
( ) - Two standard deviations

TABLE 18. (contd)

		% of Total Sample by Weight	Cs-137	Cs-134	Co-60	ZrNb-95	Eu-155	Ce-144	Ce-141	Ra-224	Ra-226	Ru-106	K-40
CC3-1													
	Sand	99.5	0.770(0.18)	<0.069	<0.023	<0.077	N.D.	N.D.	N.D.	N.D.	0.757(0.33)	<0.131	7.13(3.4)
	Silt	0.5	1.74(0.38)	<0.097	N.D.	<0.259	3.361(0.97)	N.D.	N.D.	N.D.	0.673(0.66)	<0.718	5.56
	Clay	0.003	<22.3	<18.8	<1.22	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	<17.1
CC3-2													
	Sand	98.7	0.619(0.073)	<0.017	N.D.	<0.031	<0.364	<0.158	<0.019	<0.042	0.512(0.12)	<0.010	5.72(1.3)
	Silt	1.3	1.26(0.13)	<0.030	<0.007	N.D.	N.D.	N.D.	N.D.	0.677(0.19)	0.696(0.20)	<0.016	7.62(2.0)
	Clay	0.004	<14.0	<0.473	<0.570	<1.45	94.1(60)	N.D.	N.D.	N.D.	<0.748	<3.58	<6.65
CC3-3													
	Sand	99.8	1.31(0.26)	<0.034	N.D.	N.D.	N.D.	N.D.	N.D.	0.945(0.45)	0.890(0.45)	N.D.	8.89(4.6)
	Silt	0.2	2.51(0.67)	<0.071	N.D.	<0.428	9.29(1.8)	N.D.	N.D.	N.D.	<1.01	<1.23	<6.55
	Clay	0.003	<4.83	<0.391	<0.425	<0.098	N.D.	N.D.	N.D.	N.D.	<0.515	<0.594	<50.4
CC5-1													
	Sand	89.0	0.250(0.029)	<0.013	N.D.	<0.023	N.D.	0.178(0.12)	<0.024	0.183(0.049)	0.350(0.05)	<0.035	3.46(0.54)
	Silt	10.7	0.312(0.029)	<0.014	<0.006	N.D.	N.D.	0.177(0.11)	0.065(0.047)	0.482(0.047)	0.414(0.05)	<0.052	4.00(0.52)
	Clay	0.3	4.86(1.0)	<0.104	<0.234	<0.289	7.88(2.5)	N.D.	N.D.	N.D.	<1.57	<2.29	36.8(19)
CC5-2													
	Sand	81.0	0.955(0.05)	0.033(0.02)	<0.026	<0.007	<0.059	N.A.	N.A.	0.688(0.06)	0.601(0.06)	<0.053	11.8(0.74)
	Silt	18.7	0.529(0.04)	0.033(0.02)	N.D.	N.D.	N.D.	N.A.	N.A.	0.803(0.05)	0.751(0.06)	<0.017	8.13(0.63)
	Clay	0.3	7.38(1.1)	<0.407	N.D.	N.D.	N.D.	N.A.	N.A.	<1.38	1.86(1.8)	N.D.	37.1(19)
	Organic		1.72(0.61)	<0.104	<0.224	<0.406	N.D.	N.A.	N.A.	N.D.	<0.405	4.93(4.1)	<9.49
CC-9													
	Sand	98.9	0.239(0.058)	<0.012	N.D.	N.D.	N.D.	<0.094	<0.020	0.354(0.11)	0.478(0.11)	<0.061	5.60(1.2)
	Silt	1.1	0.706(0.13)	N.D.	<0.019	<0.053	N.D.	<0.106	<0.048	0.975(0.23)	0.874(0.24)	N.D.	8.56(2.4)
	Clay	0.1	<1.186	<0.040	<0.002	N.D.	N.D.	N.D.	N.D.	N.D.	<0.053	N.D.	<21.4
CC-11													
	Sand	87.4	80.6(0.33)	1.44(0.05)	0.854(0.05)	N.D.	0.633(0.07)	N.A.	N.A.	1.31(0.05)	0.756(0.05)	0.344(0.14)	14.5(0.68)
	Silt	11.3	21.4(0.32)	0.445(0.08)	0.308(0.08)	N.D.	N.D.	N.A.	N.A.	N.D.	1.59(0.16)	N.D.	12.1(1.6)
	Clay	1.3	113(2.2)	2.17(0.64)	<0.246	N.D.	N.D.	N.A.	N.A.	<0.292	1.41(1.1)	<1.11	39.7(12)

N.D. - Not detected
N.A. - Not analyzed
( ) - Two standard deviations

TABLE 19. Gamma Ray Spectrometry - Radionuclides Dissolved in Water (pCi/ℓ)

		Cs-137	Cs-134	Co-60	ZrNb-95	Eu-155	Ce-144	Ce-141	Ra-224	Ra-226	Ru-106	K-40
FC-1	(1)	0.12 (0.10)	N.A.	N.D.	N.D.	N.D.	N.D.	N.A.	N.A.	N.D.	N.D.	N.D.
	(2)	0.194 (0.02)	0.005 (.002)	0.044 (.004)	N.A.	N.A.	N.A.	N.A.	N.A.	N.A.	0.527 (.05)	N.A.
BC-3		<.040	0.040 (.033)	<.0123	0.069 (.050)	<.050	N.D.	<.240	N.D.	<.064	<.210	6.97 (4.12)
BC-4	(1)	N.D.	N.A.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	0.42 (.15)	N.D.	N.D.
CC-1		0.122 (.104)	0.084 (.07)	<.074	0.092 (.065)	0.110 (.097)	<.119	<.009	N.D.	<.075	<.095	4.53 (2.72)
CC-3		<.028	0.049 (.032)	<.018	0.116 (.106)	0.263 (.234)	N.D.	<.064	N.D.	<.080	<.095	3.91 (.60)
CC-5		0.026 (.022)	<.083	<.003	0.048 (.037)	0.067 (.06)	<.059	<.026	N.D.	<.041	<.231	3.12 (2.0)
CC-9		<.049	0.079 (.063)	<.017	0.141 (.108)	0.172 (.063)	<.146	<.021	N.D.	0.114 (.063)	<.102	4.23 (2.67)

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(1) University of Washington
(2) PNL

Remaining samples analyzed by U.S. Testing

N.D. Not Detected
N.A. Not Analyzed
( ) Two Standard Deviations

TABLE 20. Gamma Ray Spectrometry - Radionuclides Dissolved in Water (pCi/)

	Resin Beds			Aluminum Oxide Beds				
	R-1	R-2	R-3	Al-1	Al-2	Al-3	Filters	Total Activity
BC-3								
Ru 106	<.210	<.150	<.00632	<.151	<.084	N.D.	<.0127	less than
Ra 226	<.0636	<.0195	<.00138	<.0415	<.0168	<.0268	<.0582	less than
Cs 137	<.0403	<.0144	<.0004	<.0274	<.0121	<0.246	<.00171	less than
Cs 134	N.D.	<.0208	<.0020	<.0227	N.D.	N.D.	0.040 (.033)	0.040
K 40	2.86 (1.2)	<.263	0.882 (.605)	<.709	<.814	1.33 (1.1)	1.90 (0.73)	6.97
ZrNb 95	N.D.	<.0397	N.D.	N.D.	N.D.	<.0285	0.0686 (.050)	0.0686
Co 60	N.D.	<.0123	<0.0144	N.D.	N.D.	<.0016	N.D.	less than
Ce 141	N.D.	N.D.	N.D.	<.00905	<.240	<.0207	<.0285	less than
Eu 155	N.D.	N.D.	N.D.	N.D.	N.D.	<.0199	<.0505	less than
CC-1								
ZrNb 95	0.0758 (.06)	<.0473	<.529	<.0512	.0475	<.0714	0.0920 (.065)	0.1678
Eu 155	<.00136	<.0225	N.D.	<.0330	.0671	<.0823	0.110 (.097)	0.110
Ce 144	<.109	N.D.	N.D.	<.199	N.D.	N.D.	N.D.	less than
Ra 226	<.00703	<.0747	<.0232	N.D.	N.D.	<.00108	<.0348	less than
Cs 137	<.00931	<.00311	0.0458 (.036)	<.00763	.0330	0.0758 (.0703)	<.0200	0.1216
Cs 134	<.0384	<.0323	<.00379	<.0192	.0135	0.0839 (.0703)	<.0333	0.0839
Co 60	<.0176	N.D.	N.D.	<.0119	N.D.	<.0736	N.D.	less than
K 40	1.16 (.81)	N.D.	1.05 (.76)	<1.34	1.06	<.844	2.32 (.88)	3.48
Ce 141	N.D.	N.D.	<.00467	<.00866	N.D.	N.D.	N.D.	less than
Ru 106	N.D.	N.D.	N.D.	<.0181	.0952	<.0271	N.D.	less than
CC 3-3								
ZrNb 95	0.058 (.053)	<.0288	.585 (.053)	<.0862	.0585		<.0350	0.1165
Eu 155	0.0835 (.079)	N.D.	<.0492	0.180 (.15)	.00184		<.0176	0.2635
Cs 137	<.00506	<.0106	N.D.	N.D.	.00985		<.0273	less than
Cs 134	0.0488 (.032)	<.00318	<.0101	<.0406	.0479		<.00413	0.0488
Co 60	<.0179	<.00756	<.0129	N.D.	.00558		<.0116	less than
K 40	<.433	0.870 (.062)	0.668 (.062)	<.998	.945		2.37 (.703)	3.908
Ce 141	N.D.	<.0022	<.0140	N.D.	.0637		<.0184	less than
Ru 106	N.D.	<.0950	N.D.	<.0323	.141		N.D.	less than
Ra 226	N.D.	<.0255	N.D.	<.0800	.07965		<.0276	less than

TABLE 20. (contd)

	Resin Beds			Aluminum Oxide Beds				
	R-1	R-2	R-3	Al-1	Al-2	Al-3	Filters	Total Activity
CC-5								
ZrNb 95	<.0283	N.D.	0.0478 (.037)	N.D.	<.0404	<.0658	<.0252	0.0478
Eu 155	<.0545	N.D.	0.0668 (.06)	<.0555	N.D.	<.0869	N.D.	0.0668
Ce 141	<.0259	N.D.	<.0118	<.0088	N.D.	<.0071	N.D.	less than
Ra 226	<.0213	N.D.	<.0318	<.0178	<.0408	<.0303	<.0266	less than
Cs 137	<.0134	<.0191	<.0096	<.0037	<.0161	<.0072	0.0260 (.022)	0.0260
Cs 134	<.0826	<.0203	<.0019	<.0092	<.0033	<.0282	<.0136	less than
K 40	<.384	0.582 (.44)	<.175	<.775	0.949 (.81)	<.0033	1.59 (.50)	3.12
Ce 144	N.D.	<.0588	N.D.	N.D.	N.D.	N.D.	N.D.	less than
Co 60	N.D.	<.0061	<.0009	N.D.	N.D.	<.0022	<.0026	less than
Ru 106	N.D.	N.D.	N.D.	<.231	N.D.	<.108	<.0555	less than
CC-9								
ZrNb 95	0.0607 (0.53)	0.0799 (.053)	N.D.	<.0187	N.D.	<.0949	<.0067	0.1406
Eu 155	<.0809	<.0720	N.D.	<.0254	<.0410	0.172 (.065)	N.D.	0.172
Ce 144	<.146	<.0650	<.0304	N.D.	N.D.	N.D.	N.D.	less than
Ce 141	<.0206	N.D.	N.D.	N.D.	N.D.	<.0006	<.0204	less than
Cs 137	<.0203	<.0039	<.0146	<.0237	<.0013	<.0492	<.0143	less than
Cs 134	<.0316	<.0259	<.0150	<.0103	<.0016	0.0791 (.063)	<.0213	0.0791
Co 60	<.0169	N.D.	<.0088	N.D.	<.0024	<.0021	N.D.	less than
K 40	0.881 (.722)	<.496	0.857 (.674)	<.612	<.0689	<.243	2.49 (.722)	4.23
Ru 106	N.D.	<.0024	<.0006	N.D.	<.0329	<.0255	<.1020	less than
Ra 226	N.D.	<.0010	<.0100	<.0101	<.0238	<.0322	0.114 (.063)	0.114

N.D. Not detected

( ) ± two standard deviations

R-1 through R-3 Resin Beds

Al-1 through Al-3 Aluminum Oxide Beds

TABLE 21. Additional Gamma Ray Spectrometry Analysis, University of Washington

	Suspended Sand pCi/g	Suspended Silt pCi/g	Suspended Clay pCi/g	Bed Sand pCi/g	Bed Silt pCi/g	Bed Clay pCi/g	Water pCi/ℓ
FC-1							
Cs-137	10.5 (2.1)	5.17 (0.26)	11.3 (0.6)	1.04 (0.11)	0.36 (0.09)	1.11 (0.11)	0.12 (0.10)
Nb-95						0.47 (0.39)	
Ce-144						0.58 (0.32)	
Ra-226		1.04 (0.11)	1.17 (0.24)	0.51 (0.08)	0.87 (0.10)	1.53 (0.11)	
Ru-106						1.33 (0.69)	
Rh-101						0.05 (0.03)	
Sb-125						0.23 (0.15)	
Pb-210					4.0 (3.6)		
K-40		16.9 (2.8)	42.0 (6.6)	9.4 (1.9)	15.0 (2.7)	34.6 (3.1)	
Th-232		1.19 (0.35)	2.19 (0.96)	0.64 (0.28)	1.13 (0.34)	2.39 (0.32)	
Th-228		1.17 (0.13)	1.80 (0.29)	0.53 (0.09)	0.86 (0.12)	1.90 (0.14)	
U-235		0.17 (0.07)	0.40 (0.19)	0.06 (0.05)	0.11 (0.16)	0.17 (0.09)	
U-238		2.40 (0.88)	4.1 (2.2)	1.5 (0.7)	1.72 (0.92)	2.4 (1.0)	
BC-1							
Cs-137						0.17 (0.16)	
Ce-144					0.70 (0.52)		
Ra-226		0.99 (0.15)	1.32 (0.35)	0.61 (0.07)	1.33 (0.17)	1.61 (0.24)	0.42 (0.15)
Ru-103						2.3 (1.8)	
Ru-106					2.02 (1.23)		
Rh-101					0.08 (0.05)		
K-40	37.5 (31.9)	19.1 (4.2)	39.5 (9.6)	9.6 (1.9)	19.7 (4.2)	32.6 (6.5)	
Th-232		1.12 (0.60)	2.42 (1.26)	0.47 (0.25)	2.09 (0.61)	1.93 (0.75)	1.27 (0.76)
Th-228		1.03 (0.20)	2.55 (0.47)	0.49 (0.09)	1.43 (0.21)	2.07 (0.31)	
U-235		0.22 (0.12)		0.10 (0.5)	0.18 (0.13)	0.27 (0.21)	
U-238		3.1 (1.3)	4.1 (3.3)		2.0 (1.5)	2.75 (0.25)	

TABLE 21. (contd)

	BC-4 Mixed Bed pCi/g	CC-3 Mixed Bed pCi/g	CC-11 Mixed Bed pCi/g
Cs-137		1.33 (0.13)	101.0 (1.0)
Co-60	0.10 (0.06)		0.79 (0.09)
Eu-152			0.25 (0.13)
Ra-226	0.39 (0.11)	0.46 (0.08)	0.89 (0.22)
Rh-101	0.05 (0.04)		
Rh-102			0.45 (0.18)
Sb-124			1.49 (0.43)
Bi-207			0.16 (0.11)
K-40		11.5 (2.1)	15.5 (2.3)
Th-232	0.75 (0.29)	0.65 (0.19)	1.0 (0.8)
Th-228	0.48 (0.11)	0.65 (0.09)	1.0 (0.2)
U-238	0.99 (0.56)	0.96 (1.4)	

A blank indicates that the radionuclide actually was below detection

( ) - ± standard deviation for counting error the detection limits for the small sample sizes vary significantly, resulting in many "less than" values in Table 17. However, because of the extremely small percentages of sand and clay size fractions most of the radioactivity of the total (composite) suspended sediment sample would be contained in the silt size fraction.

Cesium-137 was the principal gamma radionuclide whose activity levels in samples downstream of the NFS complex were consistently higher than the activity in the two upstream control samples. Cesium-134 and ^{60}Co levels in suspended sediment of FC-1 were higher than the levels in upstream control transects, but at downstream transects the levels were generally indistinguishable from background levels. At BC-1, the upstream control transect on Buttermilk Creek, ^{137}Cs levels associated with suspended sand, silt and clay were, respectively, 0.63 pCi/g, 0.11 pCi/g, and 0.36 pCi/g. At CC-1, the upstream control transect on Cattaraugus Creek, ^{137}Cs levels associated with the suspended sand and suspended clay fractions were less than the counting error.

Figure 8 is a comparison of ^{137}Cs levels associated with the suspended sand, silt and clay fractions of each sample. The highest ^{137}Cs levels in suspended sediments were generally found in Buttermilk Creek and the one Franks Creek sample, and were associated with the suspended sand and clay fractions. At BC-2, station 2 and BC-3, station 2, the ^{137}Cs levels associated with suspended sand were 14.50 ± 0.82 pCi/g and 13.6 ± 1.1 pCi/g, respectively. These two transects are the first two transects downstream of Franks Creek which drains the NFS complex. Cesium-137 associated with suspended sand at FC-1 (10.5 ± 2.1 pCi/g) was slightly lower than BC-2, station 2 and BC-3, station 2, however, the suspended silt and clay fractions with ^{137}Cs values of 5.17 ± 0.26 pCi/g and 11.3 ± 0.6 pCi/g, respectively, at FC-1 were the highest levels associated with the suspended silt and clay fractions found during the Phase 1 sampling program.

The ^{137}Cs levels in suspended sediment of Cattaraugus Creek were generally lower than those in Buttermilk and Franks Creeks, and in many cases the levels were comparable to the levels in suspended sediment of the two upstream control stations. In a few cases, such as at CC-3, Felton Bridge, CC-5 Springville Reservoir and CC-11, mouth of Cattaraugus Creek, elevated levels of ^{137}Cs were found associated with one or more size fractions of

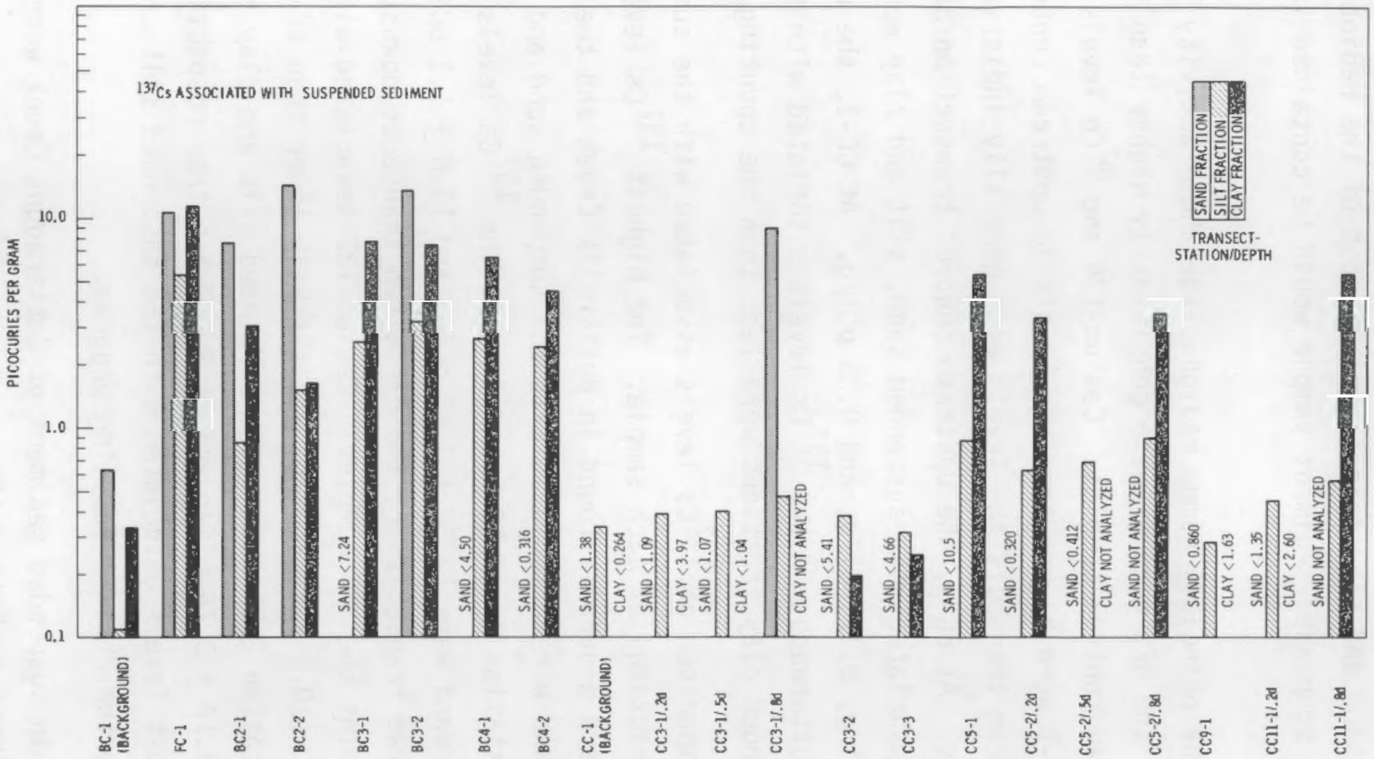

FIGURE 8. Cesium-137 Associated with Suspended Sediment

suspended sediment. Near the bed of CC-3, station 1 the suspended sand fraction had a ^{137}Cs value of 9.02 ± 2.2 pCi/g. All the suspended clay fractions of samples taken in Springville Reservoir, except for CC5-2/.5d, had ^{137}Cs levels slightly greater than 5 pCi/g. The suspended clay fraction near the bed at the mouth of Cattaraugus Creek (CC11-1/.8d) had a ^{137}Cs value of 5.50 ± 4.7 pCi/g.

There was some lateral variability and depth variability of ^{137}Cs associated with suspended sediment at some transects. At BC-2, stations 1 and 2 (station locations shown in Appendix C), for example, the ^{137}Cs levels associated with suspended sand varied from 7.6 ± 0.47 pCi/g to 14.50 ± 0.80 pCi/g, respectively. If the composite ^{137}Cs activity of suspended sediment is considered, however, the lateral variability is much smaller (2.17 pCi/g vs 2.71 pCi/g) at BC-2. Where multiple depth samples were taken (CC3-1, CC5-2, CC11-1) the ^{137}Cs levels in suspended sediment were generally higher near the bed than near the water surface.

Gamma Activity Associated with Bed Sediment

Table 18 and portions of Table 21 summarize the results of gamma ray spectrometry of bed sediment. In contrast to suspended sediment samples collected during the Phase 1 sampling program, the bed sediment was comprised almost entirely of sand size (or greater) material. Seldom was the sand fraction less than 90% of the total bed material. Only in areas where the flow velocities were small, such as at the mouth of Franks Creek (FC-1), Springville Reservoir (CC-5), and mouth of Cattaraugus Creek (CC-11), was the bed comprised of less than 90% sand size material. Although the gamma ray activity of the silt and clay fractions may be exceptionally high in many cases, the total bed activity may be low unless the corresponding sand fraction also has high levels.

As with the gamma ray activity associated with suspended sediment, ^{137}Cs is also the principal gamma emitter in bed sediment. The ^{134}Cs and ^{60}Co levels in bed sediment of Franks Creek (FC-1) were higher than those measured at the upstream control transects, but the levels at downstream transects on Buttermilk and Cattaraugus Creeks generally could not be distinguished from background levels. The ^{137}Cs levels at the two upstream control transects are very low, the highest background value being found associated with the bed clay fraction of CC-1 (0.16 ± 0.08 pCi/g). In very few cases were the ^{137}Cs levels in bed samples downstream of the NFS complex found to be at or below these control levels.

A comparison of ^{137}Cs in each of the bed sand, silt and clay size fractions of Phase 1 samples is given in Figure 9. The highest ^{137}Cs in bed sediments were found in Buttermilk Creek and at the mouth of Cattaraugus Creek (CC-11). The clay fraction generally had the highest ^{137}Cs levels, as would be expected, due to the large specific surface area of clay size sediment and the affinity of ^{137}Cs for sediment. An exception to this was BC-4, station 1, where the ^{137}Cs level associated with bed sand was 125 ± 7.4 pCi/g. Three samples had ^{137}Cs levels greater than 100 pCi/g. These samples were located at BC-3, station 1 (bed clay - 125 ± 3.1 pCi/g), BC-4, station 1 (bed sand - 125 ± 7.4 pCi/g), and CC-11 (bed clay - 113 ± 2.2 pCi/g).

The ^{137}Cs levels in the bed sample from Franks Creek (FC-1) was surprisingly low. The bed sand and clay fractions were 1.04 ± 0.11 pCi/g and 1.11 ± 0.11 pCi/g, respectively. The bed silt fraction had a ^{137}Cs value of 0.37 ± 0.09 pCi/g. The ^{137}Cs levels of bed sediment in the fast flowing reaches of Cattaraugus Creek, i.e., CC-3 and CC-9 were relatively lower than areas where the flow was moving slowly.

Gamma Activity Associated with Water

Tables 19, 20 and portions of Table 21 are the results of gamma ray spectrometry analysis of radionuclides dissolved in water. In Table 20 the results are reported as the gamma activity associated with each of the resin beds, aluminum oxide beds, filters and total activity. The gamma activity in water was very low during the Phase 1 sampling program. Even ^{137}Cs levels in water were difficult to distinguish from the levels in the upstream control transect on Cattaraugus Creek. The counting error as indicated by two standard deviations, in most cases, were very high, even though some of the samples were counted for 2000 minutes. These large counting errors are not only a result of the low levels of activity, but also due to the size of the

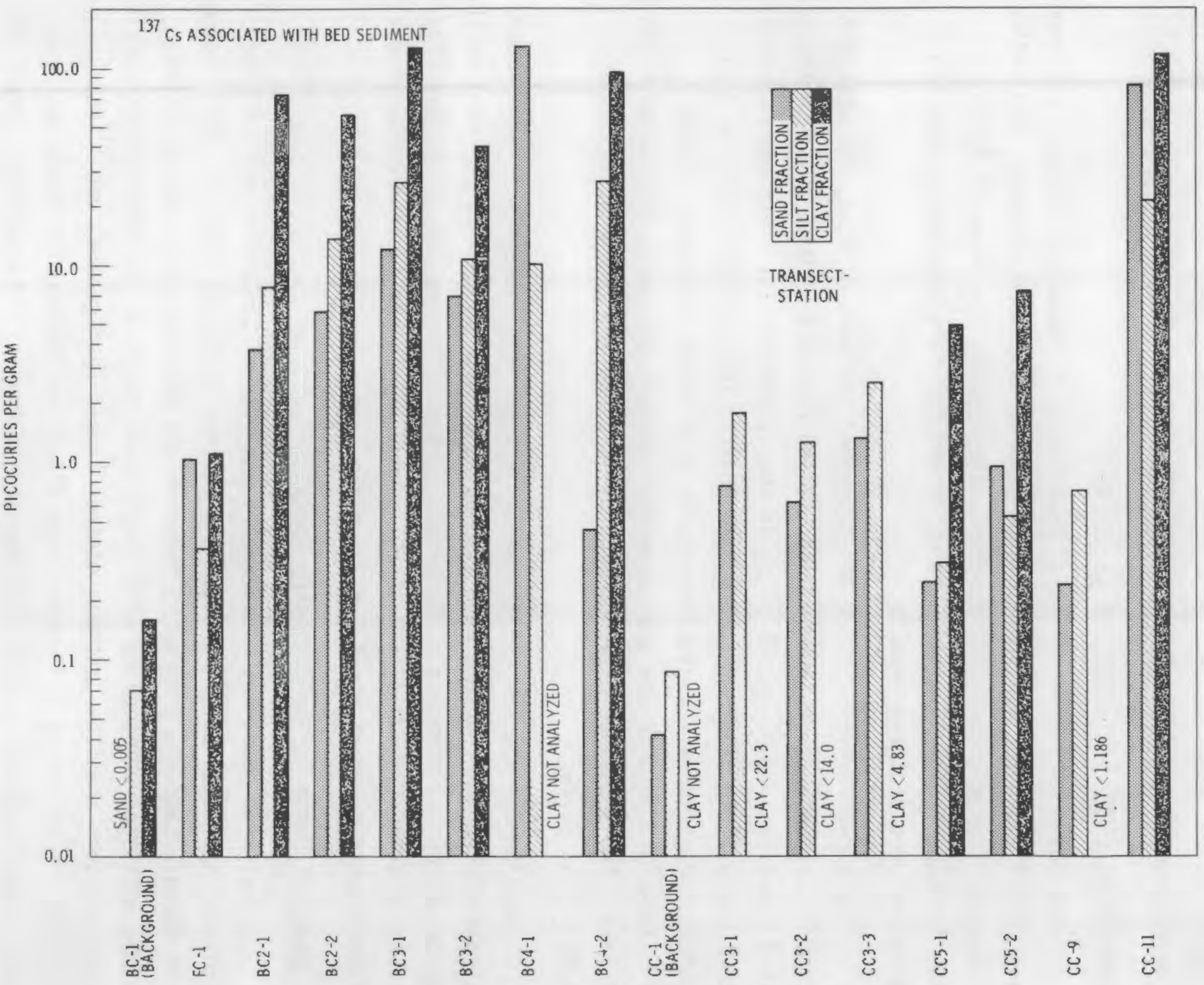

FIGURE 9. Cesium-137 Associated with Bed Sediment

water sample taken. A different method for collection of large volume water samples, used during the Phase 2 sampling program, allowed larger water samples to be taken in the field. This method will be discussed further in the section on the Phase 2 sampling program.

Additional Gamma Ray Spectrometry Analysis

The University of Washington, Department of Radiation Ecology, performed additional gamma ray spectrometry analysis of suspended sediment, bed sediment and water from a number of transects. The results are shown in Table 21. Thorium-232, ^{228}Th, ^{235}U and ^{238}U which had not been previously reported were included in these results.

Thorium-232 levels in bed and suspended sediment of BC-1, the upstream control transect on Buttermilk Creek ranged from 0.47 ± 0.25 pCi/g in the bed sand fraction to 2.42 ± 1.26 pCi/g in the suspended clay fraction. Water of BC-1 had a ^{232}Th value of 1.27 ± 0.76 pCi/ℓ. Thorium-232 levels in downstream sediments were indistinguishable from these background levels. Thorium-228 levels in bed and suspended sediment were very similar to ^{232}Th levels. The suspended clay fraction of BC-1 had a ^{228}Th value of 2.55 ± 0.47 pCi/g.

Uranium-235 and ^{238}U levels in Cattaraugus and Buttermilk Creeks, downstream of the NFS complex, were also below or near to the levels found at the upstream control transect on Buttermilk Creek. Uranium-235 in the bed clay fraction at BC-1 was 0.27 ± 0.21 pCi/g and ^{238}U in the suspended clay fraction at this transect was 4.1 ± 3.3 pCi/g.

Summary of Gamma Ray Spectrometry Analysis

Results of gamma ray spectrometry analysis of suspended sediment, bed sediment and water of Cattaraugus and Buttermilk Creeks indicate that during the Phase 1 sampling program in November and December 1977, the principal gamma emitting radionuclide with levels downstream of the NFS complex consistently higher than the upstream control levels (background) was ^{137}Cs. Although ^{134}Cs levels in Franks Creek (FC-1) and Buttermilk Creek was generally higher than background; the downstream levels in Cattaraugus Creek, in most cases, were not significantly higher than the background levels. The levels of gamma emitters other than ^{137}Cs, in a few cases, were greater in downstream samples than in background samples (^{60}Co, ^{155}Eu, ^{144}Ce, ^{106}Ru, ^{95}ZrNb); but these samples were too near the background, or the number of cases were too few in number to provide an accurate evaluation.

Cesium-137 levels were highest in bed sediment, intermediate in suspended sediment and lowest in water. In some bed sediment samples ^{137}Cs levels exceeded 100 pCi/g whereas in suspended sediment samples the highest recorded levels were less than 15 pCi/g. Cesium-137 levels in suspended sediment of Franks Creek and Buttermilk Creek were generally higher than in Cattaraugus Creek. High ^{137}Cs levels in bed sediment were not restricted to Franks Creek and Buttermilk Creek. One of the highest levels of 113 pCi/g was found in the bed clay fraction of CC-11 in Cattaraugus Creek near Lake Erie.

The distribution of ^{137}Cs between sand, silt and clay size fractions in bed sediments generally showed a trend towards increasing ^{137}Cs levels with decreasing grain size. In suspended sediment, this trend was not nearly as apparent, especially in suspended sediment of Franks and Buttermilk Creeks. Cesium-137 levels in water were very low and in only a few cases were they above detection.

PLUTONIUM AND STRONTIUM ANALYSIS

Results of ^{238}Pu, ^{239,240}Pu, and ^{90}Sr analyses of suspended sediment and bed sediment are summarized in Tables 22 and 23. Only two water samples, FC-1 and BC-4 were analyzed for Pu and Sr for reasons stated previously in the section on "Radiological Analysis." The ^{238}Pu and ^{239,240}Pu in water of Franks Creek (FC-1) were 0.0134 ± 0.0026 pCi/ℓ and 0.0020 ± 0.0010 pCi/ℓ, respectively. The levels in water of BC-4, Thomas Corners Bridge on Buttermilk Creek were 0.0154 ± 0.003 pCi/ℓ and 0.0038 ± 0.0015 pCi/ℓ, respectively.

Plutonium and Strontium in Suspended Sediment

Very few suspended sediment samples had detectable levels of ^{238}Pu, and all these were found in Franks Creek and Buttermilk Creek. The highest ^{238}Pu value was found associated with the suspended silt fraction of BC3-1

TABLE 22. Strontium and Plutonium Attached to Suspended Sediment (pCi/g)

	^{238}Pu	$^{239-240}$Pu	^{90}Sr
FC-1(UW)			
Sand	N.A.	N.A.	N.D.
Silt	0.0074 (0.0007)	0.0013 (.0004)	<0.013
Clay	0.0344 (.0028)	0.0139 (.0017)	N.D.
BC-1(UW)			
Sand	N.A.	N.A.	N.A.
Silt	0.0153 (.0040)	0.0024 (.0023)	<0.029
Clay	N.A.	N.A.	N.A.
BC2-1			
Sand	0.0262 (.023)	<.00818	0.147 (.0044)
Silt	N.A.	N.A.	0.230 (.014)
Clay	N.D.	N.A.	1.02 (.18)
BC3-1			
Sand	N.D.	<1.58	<1.68
Silt	0.236 (.16)	<.107	1.15 (.066)
Clay	N.D.	<.0042	1.18 (.38)
BC4-1			
Sand	N.D.	<.0304	1.40 (1.0)
Silt	N.D.	<.003	2.27 (.092)
Clay	N.D.	<.019	1.06 (.31)
CC3-1/.2d			
Silt	<.024	0.0988 (.049)	2.40 (.12)
Clay	N.D.	N.D.	0.819 (.61)
CC3-1/.5d			
Sand	N.D.	N.D.	0.757 (.36)
Silt	N.D.	<.00478	0.354 (.038)
Clay	N.D.	<.017	1.01 (.56)
CC3-2			
Sand	N.D.	N.D.	<.64
CC5-1			
Sand	N.D.	N.D.	10.1 (4.5)
Silt	N.D.	N.D.	2.66 (.13)
Clay	N.D.	N.D.	0.770 (.28)
CC5-2/.5d			
Sand	N.D.	N.D.	7.21 (4.5)
Silt	N.D.	<.0013	1.84 (.092)
CC9-1			
Sand	<.00235	<.0163	0.717 (.33)
Silt	N.D.	N.D.	2.17 (.11)
Clay	N.D.	N.D.	0.931 (.45)
C11-1/.8d			
Silt	N.D.	<.0056	1.55 (.077)
Clay	N.D.	<.0398	0.788 (.56)

N.A. Not Analyzed
N.D. Not Detected
( ) ± one standard deviation

TABLE 23. Strontium and Plutonium Attached to Bed Sediment (pCi/g)

	^{238}Pu	$^{239,240}Pu$	^{90}Sr
FC-1(UW)			
Sand	0.0051 (.0009)	0.0030 (.0006)	<0.042
Silt	N.A.	N.A.	<0.022
Clay	0.0028 (.0009)	0.0003 (.0010)	<0.028
BC-1 (UW)			
Sand	0.0020 (.0004)	0.0006 (.0002)	<.013
Silt	0.0613 (0.0161)	0.0239 (0.0093)	<.03
Clay	0.0051 (0.0023)	<0.0014	N.D.
BC2-1			
Sand	N.D.	N.D.	0.0721 (0.045)
Silt	N.D.	N.D.	0.315 (0.045)
Clay	N.D.	<0.680	<0.592
BC3-1			
Sand	N.D.	<0.0008	0.237 (0.045)
Silt	<0.0681	<0.0487	0.234 (0.045)
Clay	N.D.	N.D.	3.38
BC4 Mixed Bed (UW)	0.0059 (.0018)	0.0065 (.0016)	N.A.
BC4-1			
Sand	N.D.	<0.284	<0.330
Clay	N.D.	<0.210	0.243 (0.045)
CC3 Mixed Bed (UW)	0.0039 (.0011)	0.0025 (.0010)	
CC3-1			
Sand	N.D.	N.D.	<0.030
Silt	N.D.	N.D.	0.0480 (.045)
Clay	N.D.	<0.473	<4.50
CC3-2			
Sand	N.D.	N.D.	<.0420
Silt	N.D.	N.D.	<0.0150
Clay	N.D.	<.0819	<1.22
CC9-1			
Sand	N.D.	<.00687	0.0841 (0.045)
Silt	N.D.	N.D.	0.0480 (0.045)
Clay	N.D.	N.D.	<0.287

N.A. Not Analyzed
N.D. Not Detected
( ) ± one standard deviation

(0.236 ± 0.16 pCi/g). At FC-1 the suspended silt and clay fractions had ^{238}Pu levels of 0.0074 ± 0.0008 pCi/g and 0.0344 ± 0.0028 pCi/g, respectively. Detectable levels of ^{238}Pu were also found in the suspended sand fraction of BC-2, station 1 and the suspended silt fraction of BC-1.

Detectable levels of ^{239,240}Pu in suspended sediment were found only at FC-1, BC-1 and near the surface (.2d) of CC-3, station 1. The highest ^{239,240}Pu value of 0.0988 ± 0.049 pCi/g was found in the suspended sand fraction of CC-3, station 1. The suspended silt and clay fractions of FC-1 had ^{239,240}Pu levels of 0.0013 ± .0004 pCi/g and 0.0139 ± .0017 pCi/g, respectively.

Detectable levels of ^{90}Sr in suspended sediment were found at most transects. The highest recorded value was found in the suspended sand fraction of CC-5, station 1 in Springville Reservoir with a ^{90}Sr value of 10.1 ± 4.5 pCi/g.

Figure 10 is a comparison of ^{90}Sr levels in the suspended sand, silt and clay fractions of Phase 1 samples. The highest ^{90}Sr levels in suspended sediment were found in Springville Reservoir (CC-5) on Cattaraugus Creek. Strontium-90 levels in the suspended sediment of Cattaraugus Creek were as high or higher than in suspended sediment of Buttermilk and Franks Creeks.

Plutonium and Strontium in Bed Sediment

Table 23 is a summary of ^{238}Pu, ^{239,240}Pu and ^{90}Sr levels in bed sediment of Cattaraugus and Buttermilk Creeks during the Phase 1 sampling program. Very few bed sediment samples had detectable levels of ^{238}Pu and ^{239,240}Pu. The highest ^{238}Pu value was found in the bed silt fraction of BC-1 (0.0613 ± 0.0161 pCi/g). The bed sand fraction of FC-1, bed clay fraction of BC-1 and mixed bed sample of BC-4 had similar ^{238}Pu levels.

The only bed sediment samples with detectable levels of ^{239,240}Pu were found at FC-1, BC-4 (mixed bed), BC-1 and CC-3 mixed bed. The highest ^{239,240}Pu value of 0.0239 ± 0.0093 pCi/g was found in the bed silt fraction of BC-1, the upstream control transect on Buttermilk Creek. Strontium-90 levels in bed sediment varied from below detection to 0.315 ± 0.045 pCi/g. The highest value was found at BC-2, station 1 in the bed silt fraction.

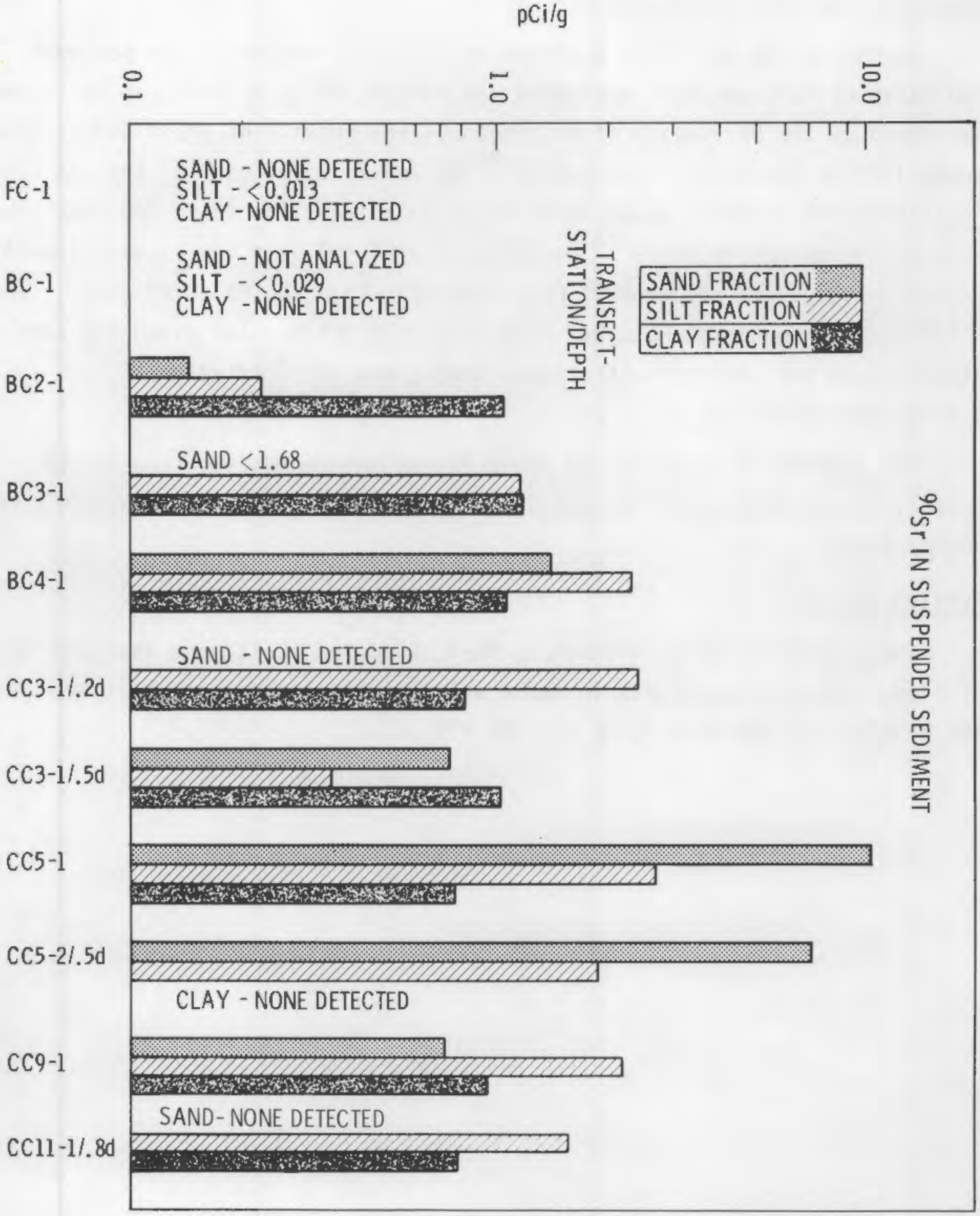

FIGURE 10. Strontium-90 in Suspended Sediment

AMERICIUM AND CURIUM ANALYSIS

Americium-241 and ^{244}Cm analysis of suspended sediment, bed sediment and water of FC-1 and BC-1 and mixed bed samples of BC-4, CC-3 and CC-11, were performed by the University of Washington. The results of these analyses are summarized in Table 24. The highest ^{241}Am levels were found in the bed clay fraction of BC-1 with a value of 1.00 ± 0.30 pCi/g. The suspended clay sample from this same sample had a ^{241}Am value of 0.94 ± 0.31 pCi/g. Americium-241 levels in suspended sediment of FC-1 were less than the counting error. The ^{241}Am value in water of FC-1 was 0.00163 ± 0.30 pCi/g. The mixed bed sample of CC-11, at the mouth of Cattaraugus Creek, had an ^{241}Am value of 0.28 ± 0.06 pCi/g.

The highest ^{244}Cm value was found in the suspended clay fraction of FC-1 (1.29 ± 0.08 pCi/g). Cesium-244 value in the water of BC-4 was 0.00004 ± .00002 pCi/ℓ.

Tritium Analysis

Three water samples, located at BC-4, CC-3 and CC-11 were analyzed for tritium. The tritium levels in these water samples were, respectively, 461 ± 35 pCi/ℓ, 305 ± 36 pCi/ℓ and 206 ±32 pCi/ℓ.

TABLE 24. Americium-241 and Curium-244 Analysis University of Washington

	Am-241 pCi/g	Cm-244 pCi/g
FC-1		
Suspended Sand	NA	NA
Suspended Silt	<0.06	0.05 (0.01)
Suspended Clay	<0.18	1.29 (0.08)
Bed Sand	0.40 (0.07)	0.28 (0.02)
Bed Silt	0.23 (0.06)	0.24 (0.02)
Bed Clay	0.74 (0 13)	0.30 (0.03)
Water (pCi/ℓ)	0.00163 (.00024)	<0.00002
BC-1		
Suspended Sand	NA	NA
Suspended Silt	0.47 (0.13)	0.50 (0.04)
Suspended Clay	0.91 (0.31)	0.64 (0.05)
Bed Sand	0.26 (0.04)	0.29 (0.03)
Bed Silt	0.28 (0.13)	0.24 (0.03)
Bed Clay	1.00 (0.30)	0.59 (0.04)
BC-4		
Mixed Bed	0.20 (0.04)	0.29 (0.03)
Water p/Ci/ℓ	<0.00046	0.00004 (.00002)
CC-3		
Mixed Bed	<0.04	<0.01
CC-11		
Mixed Bed	0.28 (0.06)	0.18 (0.02)

N.A. Not analysed due to insufficient quantity of sample

( ) ± one standard deviation

PHASE 2. SAMPLING PROGRAM

The Phase 2 field sampling program, data collection during low flow conditions, was conducted in September 1978. Results of the Phase 1 sampling program, specifically, the very low radionuclide levels found in the water of Cattaraugus and Buttermilk Creeks, has necessitated certain modifications in the sampling methodology. During the Phase 2 sampling program the volume of water processed for radiological analysis of suspended sediment and water was increased from 80 gallons to 400 gallons. The separation of suspended sediment from water was also conducted in the field in order to eliminate the need of transporting large volumes of water. The sampling apparatus used during the Phase 2 sampling program is shown in Figure 11. This method allowed the processing of a large volume of water in the field within a relatively short period of time without the necessity of returning it to the laboratory for further processing. The method has been used, in part, on the Columbia River by PNL's Physical Sciences Depatment. However, because of the higher suspended sediment load in Cattaraugus and Buttermilk Creeks, and the need to divide the particulate (suspended sediment) phase into three size fractions, the method has been extended to cover two phases - the particulate phase and dissolved phase.

The particulate phase consisted of a high-speed continuous flow centrifuge to pre-separate the suspended sediment from the water. A Westfalia Model OTA 7-00-066 Clarifuge was used. This machine has the capability of processing about 300 gallons of water per hour at about 9000 rpm. The centrifuge proved very successful in separation of suspended sediments from water during the Phase 2 sampling program. The suspended sediment retained in the centrifuge was in a form easily divided into size fractions in the laboratory.

The dissolved phase was designed to capture colloidal matter and radionuclides dissolved in water. After pre-separation of suspended sediment, the water then passed through a parallel set of three-0.3μ fiberglass filters to capture any colloidal material not retained in the centrifuge. The sediment-free water then passed through a series of three aluminum oxide beds and three

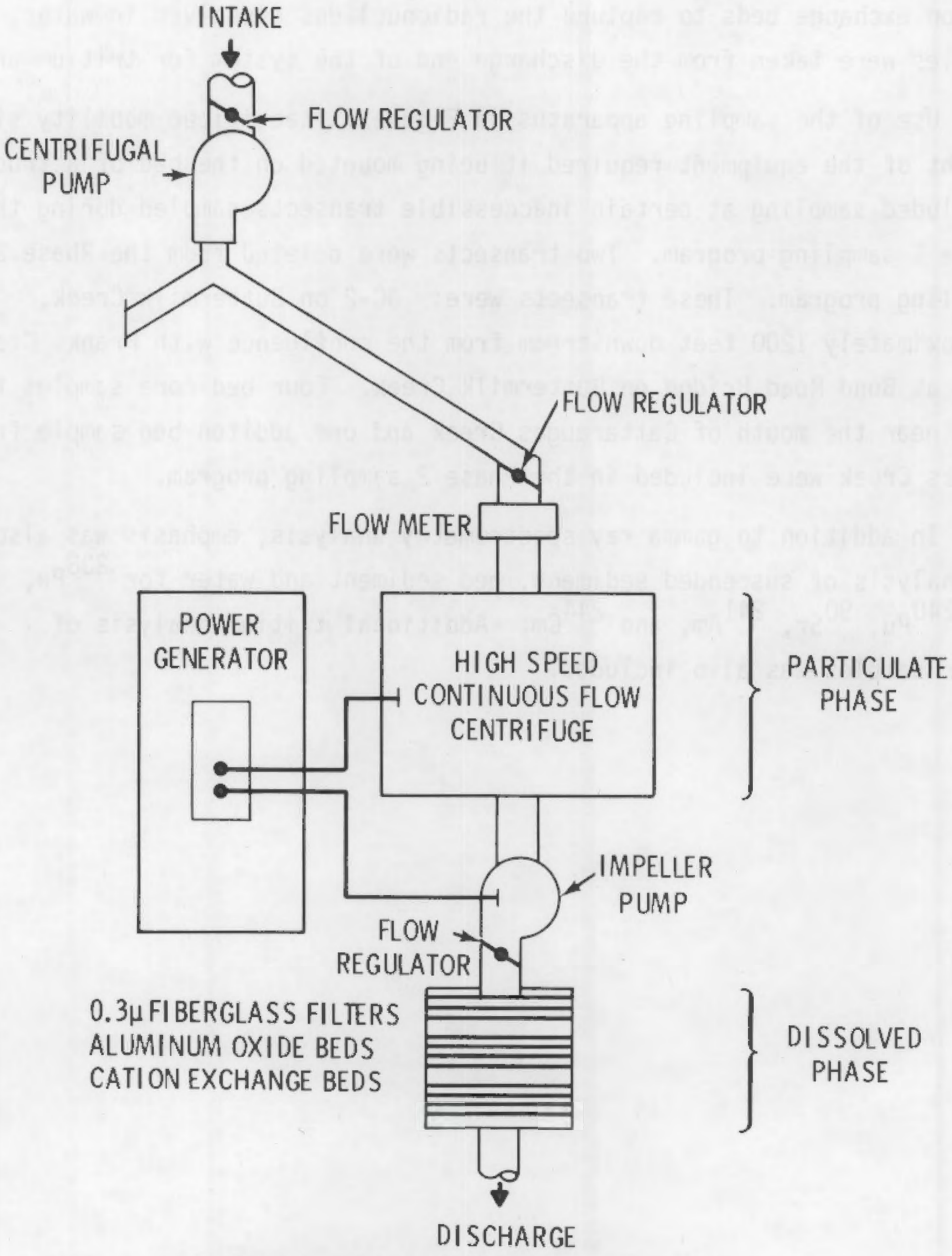

FIGURE 11. Phase 2 Sampling Procedure

cation exchange beds to capture the radionuclides dissolved in water. Water samples were taken from the discharge end of the system for tritium analysis.

Use of the sampling apparatus in Figure 11 sacrificed mobility since the weight of the equipment required it being mounted on the bed of a truck. This precluded sampling at certain inaccessible transects sampled during the Phase 1 sampling program. Two transects were deleted from the Phase 2 sampling program. These transects were: BC-2 on Buttermilk Creek, approximately 1200 feet downstream from the confluence with Franks Creek; and, BC-3 at Bond Road Bridge on Buttermilk Creek. Four bed core samples from Lake Erie near the mouth of Cattaraugus Creek and one additon bed sample from Franks Creek were included in the Phase 2 sampling program.

In addition to gamma ray spectrometry analysis, emphasis was also placed on analysis of suspended sediment, bed sediment and water for ^{238}Pu, $^{239,240}\text{Pu}$, ^{90}Sr, ^{241}Am, and ^{244}Cm. Additional tritium analysis of water samples was also included.

CONCLUSIONS

A field sampling program on Cattaraugus and Buttermilk Creeks, New York was conducted during November and December 1977 to investigate radionuclide levels in water and sediments of these two creeks for the purpose of providing data for applicaton and verification of PNL's sediment and radionuclide transport model, SERATRA. Results of radiological analysis of suspended sediment, bed sediment and water collected during the sampling program indicates a wide variability in radionuclide levels in these two creeks. Not only does this variability exist with location in Cattaraugus and Buttermilk Creeks; but also exists between water, suspended sediment and bed sediment at the same location, and between sand, silt and clay size fractions of the same sample.

Emphasis was placed on gamma ray spectrometry analysis during Phase 1 sampling program because of the large number of samples and limited funds available for radiological analysis. The principal gamma emitter in Cattaraugus and Buttermilk Creeks is ^{137}Cs. Background ^{137}Cs levels, as measured at upstream control stations on Buttermilk and Cattaraugus Creeks were less than 0.65 pCi/g in suspended sediment, less than 0.20 pCi/g in bed sediment, and less than 0.125 pCi/ℓ in water during the Phase 1 sampling program. These background ^{137}Cs levels compare favorably with background levels measured previously by other investigators both prior to and after start-up of the Nuclear Fuel Services, Inc. facility at West Valley, New York.

The highest ^{137}Cs levels in Cattaraugus and Buttermilk Creeks downstream of the NFS complex during the Phase 1 sampling program were found associated with the bed sediment. In some cases ^{137}Cs levels in the clay and sand size fractions of bed sediment exceeded 100 pCi/g. These high ^{137}Cs were found in Buttermilk Creek (BC-3 and BC-4) and at the mouth of Cattaraugus Creek (CC-11) where it enters Lake Erie. Isolated cases have been previously recorded where ^{137}Cs levels in Buttermilk Creek bed sediment exceeded 100 pCi/g. These earlier studies reported ^{137}Cs levels as the total bed activity; whereas this study reports the activity as that associated with each of the sand, silt and clay size fractions of the bed samples. The composite activity of sand, silt and clay fractions would correspond to the bulk activity reported by previous investigators. The highest ^{137}Cs levels in suspended sediments were found in Franks Creek, which drains the NFS complex, and Buttermilk Creek. Only a few suspended sediment samples had ^{137}Cs levels exceeding 10 pCi/g. Except for a few isolated cases ^{137}Cs levels associated with suspended sediment of Cattaraugus Creek were lower than Buttermilk Creek. Cesium-137 levels in the water of Cattaraugus and Buttermilk Creeks downstream of the NFS complex, during the Phase 1 sampling program, could not be distinguished from background levels.

The distribution of ^{137}Cs between the sand, silt and clay fractions in bed sediment generally reflected the affinity of ^{137}Cs for the large specific surface area of fine grained particles; e.g., the ^{137}Cs levels were lowest in the sand fraction, intermediate in the silt fraction and highest in the clay fraction. However, the distribution ratios of ^{137}Cs between the three size classes of bed sediment varied widely. The distribution of ^{137}Cs between sand, silt and clay fractions of suspended sediment did not show the trend towards increasing levels with decreasing particle size, especially in the suspended sediments of Franks Creek and Buttermilk Creek. Cesium-137 in the suspended sand fraction of these samples was often higher than the suspended silt and clay fractions.

Although in some cases gamma emitters other than ^{137}Cs were detected in suspended sediment, bed sediment and water of Cattaraugus and Buttermilk Creeks; the levels were too near the background; or the number of samples with levels above background were too few to provide an accurate evaluation. Cesium-134 levels in bed and suspended sediments of Franks Creek and Buttermilk Creek were generally above background levels. Plutonium-238, ^{239,240}Pu levels in Cattaraugus and Buttermilk Creeks during the Phase 1 sampling program were very low. The highest ^{238}Pu levels in sediments were associated with suspended silt fraction of BC-3, station 1 on Buttermilk Creek, with a value of 0.236 pCi/g. The highest ^{239,240}Pu levels in sediment were associated with the bed sand fraction of FC-1 with a value of 0.0030 pCi/g.

Strontium-90 levels associated with suspended sediment, downstream of the NFS facility, except for the one Franks Creek sample (FC-1), were consistently higher than the background levels. These higher levels, however, cannot be confidently attributed to the NFS facility because of the low levels found in Franks Creek. One suspended sand sample in Springville Reservoir (CC-5, station 1) had a level of 10.1 pCi/g. Strontium-90 levels in bed sediment and water were generally below detection.

A few ^{241}Am and ^{244}Cm analyses were performed on Phase 1 samples. The highest ^{241}Am value was found in the bed clay fraction of the upstream control transect on Buttermilk Creek (BC-1). Americium-241 in the bed clay fraction of this sample was 1.00 pCi/g.

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APPENDIX A

U.S. TESTING, INC.
Gamma Ray Spectrometry
Plutonium and Strontium Methods and Procedures

GAMMA RAY SPECTROSCOPY

GE(LI) DIODE SYSTEMS

1. Canberra Model 7220, Germanium-Lithium drifted diode. Active area facing window is 13.9 cm^2. The detector is cooled with a Canberra Model 7500 vertical dipstick cryostat and is coupled to a Canberra Model 970 spectroscopy preamplifier, a Canberra Model 1413 spectroscopy amplifier with baseline restorer and through a Canberra Model 8220A Mixer Router to one-fourth of the memory of a Canberra Model 8100 pulse height analyzer with a 4096 channel memory. Biase Voltage is supplied with a Canberra Model 3002 power supply. Data readout is accomplished with a Mohawk Data Model 2106 high speed printer. Resolution for ^{137}Cs at 662 keV is about 1.5 keV.

2. Canberra Model 7229, Germanium-Lithium drifted diode. Active area facing window is 13 cm^2. The detector is cooled with a Canberra Model 7500 vertical dipstick cryostat and is coupled to a Canberra Model 970 spectroscopy preamplifier, a Canberra Model 1413 spectroscopy amplifier with a baseline restorer and through a Canberra Model 8220A Mixer Router to one-fourth of the memory of a Canberra Model 8100 pulse height analyzer with a 4096 channel memory. Biase Voltage is supplied with a Mohawk Data Model 2106 high speed printer. Resolution for ^{137}Cs at 662 keV is about 1.5 keV.

3. Canberra Model 7248, Germanium-Lithium drifted diode. Active area facing window is 9.5 cm^2. The detector is cooled with a Canberra Model 7500 vertical dipstick cryostat and is coupled to a Canberra Model 1408C spectroscopy preamplifier, a Canberra Model 1416B spectroscopy amplifier with baseline restorer and finally to a Northern Scientific Model 600 pulse height analyzer with a 1024 channel memory. Data readout is accomplished with a Franklin high speed printer, Model 1230. Resolution for ^{137}Cs at 662 keV is about 2.8 keV.

   Instrument gain and background are determined periodically depending upon use. The systems are calibrated with radionuclide sources whose calibration values are traceable to the National Bureau of Standards. Counting efficiency checks are made daily during use. Linearity checks are made daily during use with a radioisotope whose gamma energies assure us that the system is linear. The radioisotope currently being used is ^{207}Bi.

PULSE HEIGHT ANALYSES

The Pulse Height Analyses, program for spectra from Ge(Li) Detection is written in such a manner that it can cover the whole memory of the analyzer or smaller parts of it, such as one-half or one-fourth.

The complete program is made up of two parts: the first is the calibration of the address or channels scale of the analyzer in terms of energy units. The second part of the program analyzes the spectra by locating the peaks, determining the centroid values for the peaks, calculating their corresponding energies in keV, the area for each peak and the associated standard deviation. A loop is introduced in the program to re-start the calculations for another sample, without having to go through the calibration steps for each new sample to be counted. However, the calibration step can be repeated as many times as one wishes by typing "STOP" after the analysis of a spectrum has been completed.

The areas of the peaks are calculated by summation of the number of counts in each channel of the peak and without assuming a Gaussian peak shape. If such an assumption is made, allowance should be made for asymmetry of the peaks. Kemper and van Kempen(2) used the same procedure for the same reason.

PROCESSING DATA FROM GAMMA-RAY PULSE HEIGHT ANALYZERS

A gamma-ray spectrum is obtained by plotting events detected by the analyzer on the ordinate and the pulse height on the abscissa. There is a Gaussian distribution about the gamma energy photopeak and a Compton continuum extending from zero energy to a point a little below the photopeak. Bremsstrahlung interactions add to the complexity of the lower region of the energy spectrum. Peaks may also be present from pair production and backscatter events.

Although all of a spectrum is characteristic of a particular radionuclide in a given counting geometry, only the photopeak area is currently used to identify and measure the amount of the radionuclide present. If more than one radionuclide is present, their spectra are additive.

Resolution of a complex spectrum of gamma energies into its component parts requires a determination of the Compton or other contribution from each radionuclide present to the photopeak area of each of the other radionuclides present.

For the resolution of a spectrum of "n" components, "n" simultaneous linear equations comprising "n^2" coefficients (Compton and interference correction factors) are used. If these equations are written in matrix form:

$AX = Y$ Then the solution is $X = A^{-1} Y$

Ref- A Simple Program in "Basic" Language for Analyses of Gamma-Spectra Using an On-Line Minicomputer, F. W. Lima and L. T. Atatta, Journal of Radioanalytical Chemistry, Vol. 20 (1974) 769-777

The inverse matrix A^{-1} is obtained by an iterative process performed by computer using our specially designed data processing program called "GUS". GUS uses disintegration/count conversion and Compton and interference correction factors, obtained from the calibration of crystal-analyzer-geometry configurations specified.

GUS will compute the amount of each radionuclide adjudged present and report an answer in activity per unit of sample at sampling time. At the present writing no precision estimates are included in the program. GUS requires the following input information to perform calculations:

1. Number of channels of data available
2. Counts in each channel and the channel number
3. Serial number identifying sample
4. Sampling data and time
5. Counting data and time
6. Live time of count
7. Sample size in grams or milliliters
8. Constants characteristic of the radionuclides and counting equipment
    a. Radionuclides present as a group
    b. Photopeak area or energy unique to each radionuclide
    c. Compton and interference correction factors
    d. Half-lives for decay corrections.

This information is supplied to the computer from punched paper tape and/or punched cards.

Calculation of Calibration Factors

1. Disintegration to count conversion factors, D/C.

$$D/C_{ij} = \left[\frac{D_i e^{(-\ln 2) \quad t/T_i}}{\sum_{j=a}^{b} C_{ij} - \sum_{j=a}^{b} C_{BKj}} \right]$$

where:

D_i = Disintegrations per minute for isotope "i" in counting geometry "g" at time of calibration of standard

t = Time elapsed between time of calibration of standard and time of counting

T_i = The half-life of isotope "i" in same units as "t"

$\sum_{j=a}^{b} C_{ij}$ = The sum of the counts per minute in "j" channels, "a" through "b", for isotope "i"

$\sum_{j=a}^{b} C_{BKj}$ = The sum of the background counts per minute in "j" channels, "a" through "b", where "a" and "b" are identifical for Cij and C_{BKj}

g = The geometry (sample configuration, crystal, and analyzer) used for counting

2. Compton and interference correction factors, C/C.

$$C/Cijg = \left[\frac{\sum_{j=c}^{d} Cij - \sum_{j=c}^{d} C_{BKj}}{\sum_{j=a}^{b} Cij - \sum_{j=a}^{b} C_{BKj}} \right]$$

where

$\sum_{j=a}^{d} Cij$ = The sum of the counts per minute in channels "c" through "d" for isotope "i". Channels "c" through "d" are identifical to the channels used to calculate the D/C factor for another isotope whose photopeak residues between channels "c" and "d".

For i, j, k,...isotopes, there will be i, j, k,....compton correction factors for each D/C factor.

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STRONTIUM-90 AND PLUTONIUM IN SOIL, VEGETATION AND PRODUCE SAMPLES

PRINCIPLES AND LIMITATIONS

Plutonium and strontium are leached from soil and from vegetation ash by means of a mixture of nitric and hydrochloric acids. A known amount of ^{236}Pu tracer is added. Plutonium in the tetravalent state in 8N HNO_3 are then adsorbed on ion exchange resin. Any other actinides present do not adsorb on the resin due to their different oxidation states. Concentrated HCl is passed through the resin to remove the thorium. The plutonium is removed from the resin with a dilute HF-HNO_3 solution. After evaporation to dryness and re-dissolution, the plutonium is separated by extraction into 0.5M thenoyltrifluoracetone in benzene from an aluminum nitrate-nitric acid salting solution, and then back-extracted from this into 8N HNO_3. The solution is again evaporated to dryness, re-dissolved and the plutonium is finally electrodeposited on a stainless steel disk from an acidic ammonium chloride solution with a two ampere current. Yield and plutonium activities are determined by alpha spectrometry.

Strontium and other metal ions pass through the resin in the original 8N HNO_3 solutions. The alakline earth metals are removed from the nitric acid solution by precipitation as nitrates in high nitric acid concentration. Excess calcium is removed by washing the precipitates with acetone. Strontium, barium, and any remaining calcium are dissolved in water and reprecipitated as the nitrates with fuming nitric acid. The calcium nitrate is removed by dissolving in 14.6N HNO_3. Barium is separated by precipitation as the chromate. The strontium is reprecipitated as the carbonate and beta counted with a low background beta proportional counter. Three different counts are made to determine strontium activity. If ^{89}Sr is determined to be present, an yttrium separation will confirm its presence.

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GENERAL PROCEDURES FOR STRONTIUM DETERMINATION

PRINCIPLES AND LIMITATIONS

Strontium, calcium, and barium are separated from many cations by precipitating them as the carbonates. The precipitate is dissolved in acid and the alkaline earths are precipitated as the nitrates with fuming nitric acid. Calcium nitrate is removed by dissolving with 14.6N HNO_3. Barium is separated by precipitation as the chromate. This chromate precipitation also serves to remove radium, lead, and other interferences which have carried through the fuming nitric acid precipitation. Strontium is reprecipitated as the carbonate and beta counted with a low beta proportional counter. When activity is present, at least one more count is made to determine radiochemical purity.

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APPENDIX B

UNIVERSITY OF WASHINGTON
LABORATORY OF RADIATION ECOLOGY

GAMMA RAY SPECTROMETRY, PLUTONIUM, STRONTIUM, AMERICIUM AND CURIUM METHODS AND PROCEDURES

UNIVERSITY OF WASHINGTON
LABORATORY OF RADIATION ECOLOGY

SUMMARY OF QUALITY CONTROL RESULTS OF RADIONUCLIDE ANALYSIS

INTRODUCTION

During the time period 1971-1979 the Laboratory of Radiation Ecology (LRE) has participated in internal, external, national, and international programs to compare measurements of radionuclides and stable elements. Standards as well as environmental samples have been interchanged between several laboratories including LRE and the results are reported here.

We have measured and reported about 160 intercomparison samples on about 20 radionuclides. The analysis included: gamma radionuclides by Ge(Li) diode and NaI(Tℓ) crystal methods , alpha radionuclides (by alpha spectroscopy methods for $^{238,239}Pu$, ^{241}Am, ^{210}Pb, and ZnS screen and phototube counting for gross alpha radionuclides), beta radionuclides (by radiochemistry methods for ^{90}Sr, ^{131}I, by liquid scintillation method for tritium and by low background gas counting for gross beta radionuclides), and x-ray radionuclides (by radiochemistry methods for ^{55}Fe and x-ray proportional counting); measurements of trace elements have been made by NAA and AAS methods.

It has been our policy to treat the incoming standard samples identical to incoming normal environmental samples so that our internal reliability could also be checked. No special precautions have been taken in the measurement of the quality control samples.

MEASUREMENTS OF GAMMA-RAY EMITTING RADIONUCLIDES

Measurements of the concentrations of ^{241}Am and other gamma-emitting radionuclides in samples have been made using a 1 cm^2 Ge (intrinsic)(a) detector coupled to a 400-channel pulse height analyzer for ^{241}Am and two 7.3%(b) Ge(Li)(c) detector systems coupled to two 4096 channel pulse height analyzers with a PDP-5 computer data processing and reduction system. These detection systems have been cross calibrated with the two 5 x 5 Na(Tℓ) crystal detector systems which were used previously. In addition to the cross calibration between instruments, interlaboratory calibration of samples have been made continuously over the years to insure reliability in our measurements.

(a) Applied Detector Corporation, Menlo Park, California
(b) Absolute detection efficiency for 1.33 McV gamma rays relative to a 30% efficient NaI(Tℓ) detector
(c) Nuclear Diodes, Inc., Prairie View, Illinois (presently Edax International)

The absolute counting efficiency of each instrument was determined as a function of γ-ray energy by counting a series of standards prepared in the same geometry as that used to count the samples. Each standard was prepared and contained a known amount of a given radionuclide; these standard solutions were obtained from the N.B.S. or a commercial supplier, usually Amersham. An aliquot of each standard solution was added to an acrylic casting resin and homogenized by stirring until the resin set. Each encapsulated standard was thus uniformly distributed in the volume of the counting container (2' x 1/2", 2" x 1", 3" x 2") at a standard density of 1.1 g/cc and was a "permanent" standard for future calibration checks. The results of these calibrations are shown in Figure B-1 which shows the detector efficiencies as a function of gamma energy.

Since the cpm to dpm conversion factor, which was needed to calculate the absolute radionuclide concentrations of the sediment, biota and water samples from the counting data, was a function of several variables; (e.g., gamma-ray energy and bulk density) standards were prepared at a bulk density of 1.35 by adding NaCl to increase the density of the acrylic casting resin from 1.1 to cover the range normally found in our samples. The appropriate conversion factor for each sample (density) was then approximated by linear interpolation, between the values found for the 1.1 and 1.35 g/cc density standards.

The error that could result due to possible variation of the linear dependence assumption described above was estimated by considering the case where density changes gave logarithmic rather than linear changes in the correction factor. The maximum error that could result from a logarithmic instead of the assumed linear dependence was estimated by measuring the difference in the value of the two correction factors in samples which were at the extremes of sample densities encountered (0.6 and 1.6 g/cc). The difference found using the two correction factors was 7.3% for the sample geometry and density limit of the lowest energy radionuclide of ^{241}Am (most sensitive test). For radionuclide concentrations which were determined by using higher energy gamma-rays (>59.5 Kev) and for the majority of samples which were not at the extreme limits of the densities, the error which would arise due to this uncertainty was smaller than 7.3%.

The abundance of each γ-ray observed in the spectrum was used to calculate the concentration of the radionuclide present using a weighted mean concentration of each gamma peak and its associated error (Stevenson 1966). The error term associated with the counting are 2 S.D. errors based on propagated counting statistics.

The results of interlaboratory comparisons of concentration of the gamma-emitting radionuclides in the standards and environmental samples measured are shown in Table B-1.

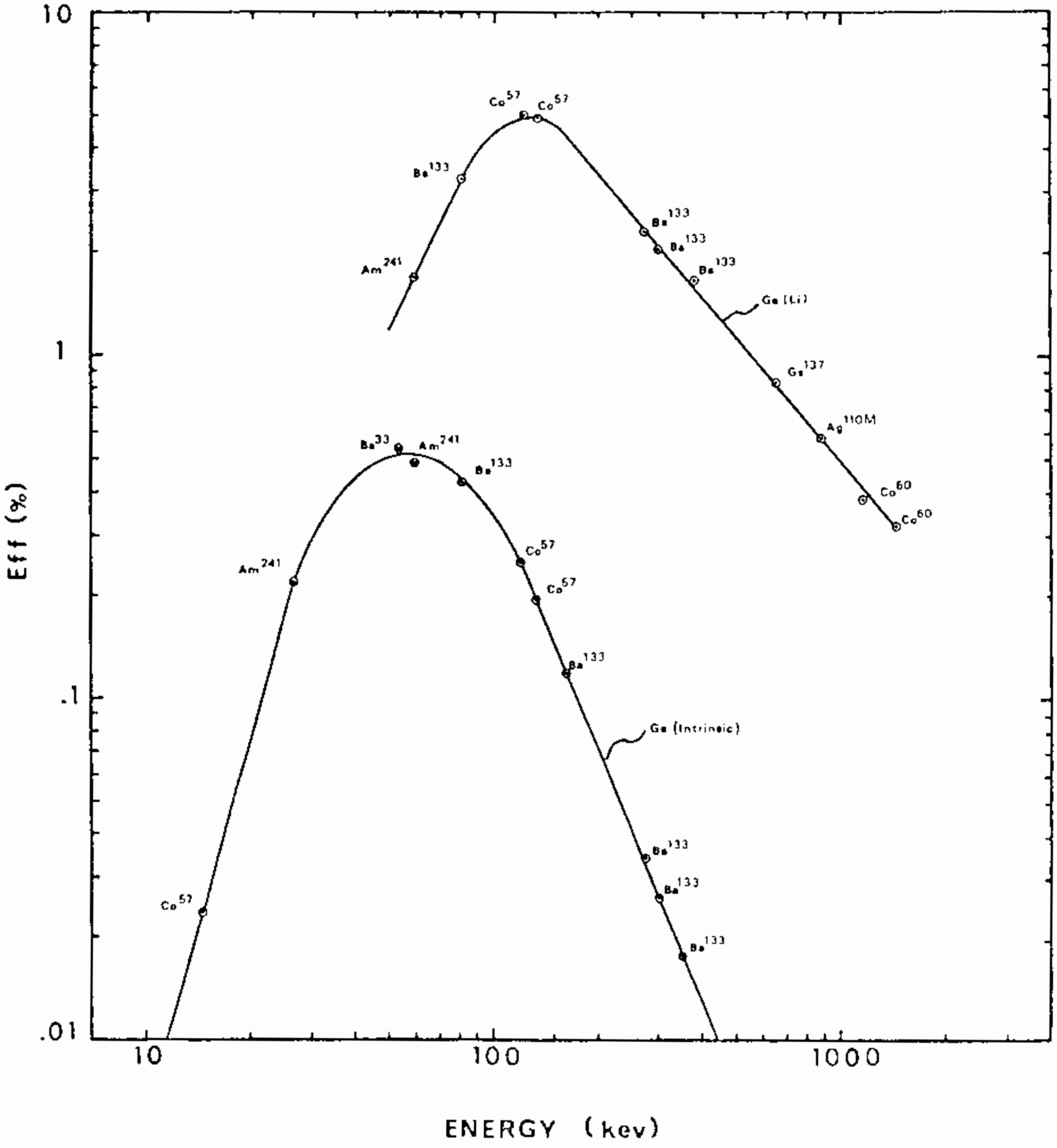

FIGURE B-1. Absolute Counting Efficiency of the Ge(Li) and Ge(Intrinsic) Detectors with Gamma-Ray Energy as Determined by Counting Radionuclide Standards Made to a Sample Density of 1.1 g/cc

TABLE B-1. Results of Interlaboratory Comparisons of Gamma and Beta Radionuclides in Samples

Sample	Type	Date	Lab	^{90}Sr	^{95}Zr-^{95}Nb	^{106}Ru	^{134}Cs	^{137}Cs
35065 MA B-1	Clam Homogenate	June 76	IAEA	7.4 ± .8		4.3 ± .6	.29 ± .06	16.24 ± .4
			LRE	6.5 ± .3		5 ± 1	.25 ± .07	18.0 ± .4
35066 MA B-2	Sea Hare	June 76	IAEA	.31 ± .09			3.0 ± 1.4	4.1 ± .3
			LRE					4.8 ± .3
35052 Milk A-9	Milk	June 76	IAEA	.49 ± .02				2.34 ± .09
			LRE	.48 ± .04				2.40 ± .3
35030 AG-I-1	Seaweed	May 72	IAEA	10.0 ± .1	2.5 ± .3	137 ± 6	10.3 ± .3	75 ± 1.
			LRE		2.21 ± .23	126 ± 4	8.5 ± .1	65.7 ± .2
35035 SD-B-1	Sediment	Jan 73	IAEA	13.8 ± .9	-	73 ± 2	9.6 ± .3	377 ± 6
			LRE			74.4 ± 1.2	9.6 ± .2	450 ± 2
35031 SW-I-1-19	Water	Jan 71	IAEA				1.7 ± .4	14.0 ± 2.0
			LRE				1.1 ± .5	16.6 ± 1.0
35032 SW-I-2-19	Water	Jan 71	IAEA			40 ± 5	26 ± 3	195 ± 14
			LRE			35.0 ± 5	28.2 ± 1.1	232 ± 4
35083 W-1	Water	1975	IAEA	3.55 ± .07				8.15 ± .12
			LRE	3.89 ± .15				7.6 ± .4
35084 W-2	Water	1975	IAEA	52.0 ± 1.0				119.3 ± 1.8
			LRE	54.0 ± 5				131.6 ± 3.4
35139	Diet	18 Feb 77	EPA	55 ± 8				45 ± 15
			LRE	69 ± 6				51 ± 3
35128	Diet	26 Nov 76	EPA	96 ± 14				46 ± 15
			LRE	112 ± 19				50 ± 12
35118	Diet	27 Aug 76	EPA					
			LRE	108 ± 9				31 ± 5
35075	Diet	Dec 75	EPA	125 ± 19				101 ± 15
			LRE	114 ± 23				104 ± 11
35053	Diet	Aug 75	EPA	101 ± 15				121 ± 18
			LRE	88 ± 2				119 ± 3
35038	Diet	Apr 75	EPA	150 ± 23				150 ± 23
			LRE	142 ± 4				155 ± 8
35028	Diet	Dec 74	EPA	175 ± 26				176 ± 26
			LRE	176 ± 2				193 ± 6
35018	Diet	Aug 74	EPA	198 ± 30				205 ± 31
			LRE					243 ± 6
35125	Milk	Nov 76	EPA	16 ± 5				11 ± 5
			LRE					9 ± 1
35110	Milk	Jul 76	EPA	0				75 ± 15
			LRE	5.8 ± 1				78 ± 7
35090	Milk	Mar 76	EPA	50 ± 7				25 ± 15
			LRE	59 ± 5				22 ± 2
35072	Milk	Nov 75	EPA	75 ± 11				75 ± 15
			LRE	68 ± 6				85 ± 2
35035	Milk	Mar 75	EPA	50 ± 8				
			LRE	51 ± 7				
35046	Milk	Jul 75	EPA	97 ± 14				70 ± 15
			LRE	89 ± 8				78 ± 4
35022	Milk	27 Nov 74	EPA	102 ± 15				101 ± 15
			LRE	10.9 ± .6				96 ± 8
35138	Water	4 Feb 77	EPA			151 ± 23	76 ± 15	39 ± 15
			LRE			105 ± 17	50 ± 7	29 ± 3
35154	Water	3 Jun 77	EPA			* *	*	*
			LRE			61 ± 3	38 ± 1	32 ± 2
35086	Water	20 Feb 76	EPA			336 ± 50	230 ± 35	351 ± 54
			LRE			263 ± 17	209 ± 2	358 ± 5
35124	Water	10 Oct 76	EPA					
			LRE			75 ± 17	82 ± 10	37 ± 5
35106	Water	18 Jun 76	EPA			79 ± 15	106 ± 16	53 ± 15
			LRE			27.4 ± 9	79 ± 3	40 ± 2
35067	Water	Oct 75	EPA			247 ± 37	349 ± 52	274 ± 41
			LRE			217 ± 19	325 ± 4	270 ± 5
35042	Water	Jun 75	EPA			325 ± 49	303 ± 46	378 ± 57
			LRE			293 ± 6	267 ± 7	369 ± 4
35031	Water	Feb 75	EPA				422 ± 63	472 ± 71
			LRE			<20	315 ± 7	398 ± 4
35021	Water	Oct 74	EPA			0 ± 15	481 ± 72	0 ± 15
			LRE			30	485 ± 61	< 6
	Water	May 74	EPA			0 ± 15	0 ± 15	0 ± 15
			LRE			<1.9	.2	<.2

* EPA results not yet received

TABLE B-1. (contd)

^{144}Ce	^{40}K	^{154}Eu	^{155}Eu	^{60}Co	^{110}Ag	^{226}Ra	^{3}H
2.7 ± .5							
2.8 ± .5							
				166 ± 7			
<2				177 ± 2			
17.4 ± .9	39 ± 3		1.5 ± .2	2.0 ± .1	1.5 ± .1		
14.7 ± 2.8	36 ± 1	.11 ± .01	1.5 ± .1	1.8 ± .1	1.2 ± .1		
129 ± 5							
118 ± 4	11.2 1.5	.94 ± .15	4.44 ± .2				
				51.5 ± .8		3 64 ± .3	2.54 ± .09
				43.1 ± 1			
				2.53 ± .04		53.3 ± 4.5	45.3 ± 1.6
				2.30 ± .02			
	2670 ± 401						
	2472 ± 83						
	2745 ± 412						
	2583 ± 237						
	2496 ± 332						
	2414 ± 362						
	2202 ± 200						
	2352 ± 353						
	1933 ± 200						
	2216 ± 333						
	2165 ± 313						
	2619 ± 393						
	2467 ± 115						
	2389 ± 358						
	2500 ± 200						
	1510 ± 76						
	1403 ± 106						
	1550 ± 233						
	1356 ± 66						
	1529 ± 229						
	1563 ± 790						
	1549 ± 233						
	1700 ± 192						
	1514 ± 228						
	1366 ± 150						
	1495 ± 224						
	1114 ± 158						
				45 ± 15			
				39 ± 5			
				*			
				26 ± 1			
				209 ± 31			
				200 ± 4			
				24 ± 3			
				53 ± 15			
				48			
				271 ± 41			
				273 ±			
				350 ± 53			
				324 ± 12			
				437 ± 66			
				420 ± 14			
				0 ± 15			
				<6			
				0 ± 15			
				<.3			

TABLE B-1. (contd)

^{131}I	^{241}Am	^{125}Sb	^{140}Ba	^{51}Cr	^{65}Zn
		2.8 ± .5			
81 ± 15 109 ± 24					
0 <30			39 ± 15 72 ± 10		
115 ± 29			174 ± 31		
127 ± 19 112 ± 19			0 <35		
0 <40			145 ± 22 <200		
149 ± 22 154 + 2			0 <50		
175 ± 26 200 ± 10			0 + 15 <55		
216 + 32 250 ± 10			207 ± 31 173 + 6		
85 ± 5 84 + 10			0 <20		
51 ± 15 48 + 2			0 <12		
75 ± 15 70 ± 3			0 <26		
75 + 15 13 ± 5			0 <56		
76 ± 15 116 ± 6			0 <20		
>.25			0 <38		
126 ± 19 119 ± 10			0 < 2		
				202 ± 30 173 ± 11	37 ± 15 37 ± 6
				*	*
				< 8	79 + 4
				0 <18	445 ± 67 472 ± 11
				<10	55 ± 9
				0 <6	79 ± 15 75.1
				0 <18	250 ± 38 277 +
				0 <20	327 ± 49 330 ± 6
				0 <30	472 ± 71 487 ± 4
				0 ± 15 <90	0 ± 15 <5
				349 ± 52 482 ± 30	0 ± 15 <.1

MEASUREMENTS OF BETA EMITTING RADIONUCLIDES

The beta-emitting radionuclides are measured using gas flow and liquid scintillation counting. The radionuclides which are measured in samples using the gas flow counters are ^{90}Sr and ^{131}I; radiochemical procedures for sample preparation are required. The results of these interlaboratory comparisons are shown in Table B-1.

MEASUREMENTS OF TRITIUM

The measurements for tritium in samples have been made by liquid scintillation methods using Instagel (Packard Instrument Co.) and a low background (4.6 c/m) detection system (Packard Tricarb). The mixture of water: Instagel was 8 cc. H_2O: 12 cc Instagel; these procedures were adopted from Sauzay and Schell (1971). Table B-2 shows that our tritium values are consistently within the measurement errors stated by EPA.

ALPHA SPECTROSCOPY MEASUREMENTS

Instrumentation and calibrations: the measurement of radioactivity by alpha spectroscopy was made by using eight 300 mm^2 silicon surface barrier diodes. Each of the two counting systems available for use consisted of four diodes, preamplifiers and amplifiers routed through a router-mixer to each of four 128-channel quadrants of a 512-channel multichannel analyzer (MCA). The MCA memory was dumped into both typwriter (digital) and graphical (analog) outputs after typical counting periods of 800 minutes. The detector amplifier gain was adjusted to 9 keV/channel. The resolution of the diodes (FWHM) was 20 keV or better. Background count rates of the four diodes used for plutonium and uranium analysis were 0-8 counts/800 minutes under each of the observed alpha peaks. Background count rates of the four diodes used for polonium analysis were typically 5 counts/800 minutes/peak.

The absolute disintegration rate of the isotopes of plutonium, uranium and ^{208}Po in the plated samples was determined by computing the ratio of the count rate observed for each isotope to the count rate for a secondary standard of known disintegration rate; corrections were made for background count rate, alpha particle branching ratios, and any impurities in the radiochemical spikes.

The disintegration rate of the secondary standards of plutonium was determined by similar calibrations with a standard ^{236}Pu solution supplied by the AEC Health and Safety Laboratories (HASL). The reliability of the plutonium calibration was verified by the agreement between the concentrations of plutonium found by this laboratory and those found by other laboratories in an interlaboratory standard solution of ^{239,240}Pu and ^{238}Pu concentrations measured by LRE in seaweed and sediment samples supplied by the International Atomic Energy Agency (IAEA) were also in agreement with the values recommended by the IAEA. The results of both these calibrations are shown in Table B-3.

TABLE B-2. Results of Interlaboratory Comparison of Tritium in Water Samples

Sample	Type	Date	Lab	^{3}H
35132	Water Cross Check	Dec. 76	EPA LRE	2300 ± 1049 2287 ± 65
---	Water Cross Check	Oct. 76	EPA LRE	58 ± 5 55 ± 9
---	Water Cross Check	Aug. 76	EPA LRE	3100 ± 1080 3200 ± 104
---	Water Cross Check	Apr. 76	EPA LRE	1776 ± 1024 1793 ± 42
35096	Water solution standard	May 76	EPA LRE	No values available 7.15 ± 0.26; 27.4 ± 0.08; 312.3 ± 0.14; 221.2 ± 3.1
35078	Water Cross Check	Dec. 75	EPA LRE	1002 ± 972 1000 ± 52
35050	Water Cross Check	Aug. 75	EPA LRE	3200 ± 1083 3337 ± 67
35036	Water Cross Check	Apr. 75	EPA LRE	1499 ± 1002 1540 ± 60
35026	Water Cross Check	Dec. 74	EPA LRE	3395 ± 1095 3449 ± 30
35017	Water Cross Check	Aug. 74	EPA LRE	1438 ± 933 1447 ± 74
---	Water Cross Check	May 74	EPA PRE	2673 ± 1050 2717 ± 38
35146	Water Cross Check	Apr. 77	EPA LRE	1760 ± 1023 1702 ± 41

TABLE B-3. Results of Interlaboratory Analysis of Samples for ^{239,240}Pu

A. Standard Reference materials, solutions, soils

Sample	Type	Date	Laboratory	^{239,240}Pu	Comments
35005 SD-B-1	sediment	January 73	IAEA	960 ± 30	^{238}Pu 42 ± 4
			LRE	950 ± 70	^{238}Pu (N.D.)
35000 AG-I-1	seaweed	January 72	IAEA	27000 ± 100	Pu 3800 ± 100
			LRE	23400 ±1000	Pu 3100 ± 100
35083 W-1	water	1975	IAEA	3.21 ± 0.05	
			LRE	2.8 ± 0.3	
35149 R-2	water	March 77	EPA	1110 ± 100	Round robin study
			LRE	990 ± 44	
Soil - 2	soil	January 71	EPA(avg.)	0.30	Cross check study
			LRE	0.16 ± 0.18	
Soil - 3	soil	January 71	EPA(avg.)	2.24	Cross check study
			LRE	0.51 ± 0.13	"High fired" soil
Soil - 4	soil	April 71	EPA(avg.)	1735 ±1220	Cross check study
			LRE	1547 ± 955	Nevada test soil
Soil - 5	soil	May 71	EPA(avg)	208 ± 117	Cross check study
			LRE	96 ± 54	"High fired" soil
Soil - 6	soil	June 71	EPA(avg)	18164 ±2800	Cross check study
			LRE	21433 ± 306	Pacific Islands soil
35047 NBS #4350	river sediment	1975	NBS	.038 ± .003	
			LRE	.042 ± .018	
LLL #110	std. solution	1973	LLL	1303 ± 28	
			LLL	1320 ± 20	
			LLL	1265 ± 5	
			MCL	1255 ± 15	
			MCL	1272 ± 6	
			LFE	1330 ± 27	
			LRE	1273 ± 64	
			EIC	1207 ± 54	

TABLE B-3. (contd)

B. Collection on Joint Cruises.

Sample	Type	Laboratory	^{239,240}Pu Particulate	^{239,240}Pu Total	Avg.±S.D.	Dev. %
Bikini Atoll - 1972						
Lagoon water - STA B-2	surface	LLL[a]	28 ± 2	107 ± 4	91.3±19	+17
		PRNC[a]		98 ± 7		+ 8
		LRE[b]	30 ± 2	69 ± 4		-24
" - STA.B-15	surface	LLL	4.7 ± .6	66 ± 2	49.3±16	+34
		PRNC	--	49 ± 4		-.6
		LRE	3.1 ± .2	33 ± 5		-33
" - STA B-15	29m	LLL	5.6 ± .6	60 ± 3	57.0±21	+ 5
		PRNC	--	76 ± 7		+33
		LRE	6.4 ± .1	35 ± 2		-38
" - STA B-25	surface	LLL	9.7 ± .9	79 ± 3	73.0±8.5	+ 8
		PRNC	--	67 ± 4		- 8
		LRE	--	--		
" - STA B-25	50m	LLL	--	64 ± 3	95 ±44	-33
		PRNC		127 ± 9		+33
		LRE	--	--		
" - STA B-30	surface	LLL	--	--		
		PRNC		55 ± 3	42 ±18	+30
		LRE	2.5 ± .3	29 ± 3		-30

TABLE B-3. (contd)

B. Collection on Joint Cruises (cont'd.) Sample	Type	Date	Laboratory	^{239,240}Pu Particulate	 Total	 Avg.±S.D.	Dev. %
			Bikini Atoll - 1972 (cont'd.)				
Lagoon water - STA B-30	45m		LLL	--	--		
			PRNC	--	81 ± 2		+15
			LRE	29 ± 1	60 ± 3		-15
Bomb Crater water - STA C-3	surface		LLL	10 ± 1	38 ± 1	44.0±16	-14
			PRNC	--	32 ± 1		-27
			LRE	13.6 ± .3	62 ± 2		+40
" - STA C-3	44m		LLL	22 ± 1	35 ± 2	33 ± 3	+ 9
			PRNC	--	--		
			LRE	24 ± 2	31 ± 3		- 9
" - STA C-8	surface	1972	LLL	--	47 ± 4	59 ±12	-13
			PRNC	--	68 ± 3		+25
			LRE	14.6 ± .6	48 ± 8		-11
Deep ocean water - STA D-1	300m		LLL	--	51 ± 6	28 ±32	+82
			PRNC	--	5 ± 1		-82
			LRE	--	--		
" - STA D-7	surface		LLL	--	3.5 ±0.2	3.45±.07	+ 1
			PRNC	--	--		
			LRE	0.13±0.06	3.4 ±1.2		- 1
			Eniwetak Atoll - 1972				
Lagoon water ½ mi. off Leroy	Surface		LLL		18 ± .9	15 ± 4	+20
		½ flood ½ ebb }	LRE	0.45 ±0.1	12 ± 3.5		-20
" Enewetak Dock	Surface	?	LLL	--	1.6 ± 0.2	1.43±.25	+12
		flood	LRE	0.47 ±0.1	1.25± 0.2		-12
" Japtan	Surface	?	LLL	--	2.8 ± 7	2.14±.65	+30
	Surface	flood	LRE	0.62 ±0.1	1.5 ± 0.2		-29
	Surface	ebb	LRE	1.15 ±0.2	2.14± 0.4		0

TABLE B-3. (contd)

B. Collection on Joint Cruises (cont'd.) Sample	Type	Date	Laboratory	239, 240Pu Particulate	 Total	Avg.±S.D.	Dev. %
Eniwetak Atoll - 1972 (cont'd.)							
Lagoon water - Runnit Dock	Surface	?	LLL	--	43.6 ± 1.4	57.1 ± 19	-23
		½ flood ½ ebb	LRE	26.9 ±1.4	70.6 ± 6.6		+23
" - 200 yds off Runnit	15m	?	LLL		77.0 ± 3.1	69.1 ± 11	+11
		flood	LRE	34.3 ±0.9	61.1 ± 2.6		-11
Crater water - Mike Crater	33m	?	LLL	--	1510 ± 60	844 ±941	+79
		ebb	LRE	164 ±5	179 ± 6		-79
" " "	Surface		LLL	--	19.0 ± 0.8	20.0±1.5	-5
		flood	LRE	11.13 ±0.6	21.1 ± 5.6		+5
Washington Coastal Waters - 1976							
Coastal Water JDF-8 N 48° 27.1; W 124°45.2"	Surface		BNWL	0.14 ± .01)	(0.69±0.12)	- Sequim Bay	
			LRE	< .06	0.34±0.1		
			LRE		< 0.59		
			LRE (batch)		0.5 ±0.25		
JDF-8 (50m) N 48° 30.0'; W 126° 46.0'	Surface		BNWL	0.09 ±0.01	0.12±0.04		
			LRE	<0.008	0.14±0.14		
			LRE	0.061±0.045	0.19±0.19		
			LRE (batch)		< .44		
HOH-5 mi. N 47°40'; W 124°33.6'	Surface		BNWL	0	0.18±0.05		
			LRE	0.03	0.26±0.26		
			LRE (batch)		< .4		

TABLE B-3. (contd)

C. Internal Comparisons of BLVWS and Batch Methods

				^{239,240}Pu			Dev.
Sample	Type	Depth	Method	Particulate	Total	Avg.±S.D.	%
			Bikini Atoll - 1976				
Lagoon Water	STA B-3	Surface	Batch	--	55.1 ± 17.4	48.9 ± 9	+13
			BLVWS	16.7 ± 1.0	42.7 ± 2		-13
"	"	29 m	Batch	--	72.2 ± 8.2	67.5 ± 7	+ 7
			BLVWS	50.2 ± 3.6	62.9 ± 4		- 7
"	STA B-8	Surf	Batch	--	41.8 ± 9.7	34.7 ±10	+20
			BLVWS	< .3	27.7 ± 3.7		-20
"	"	17m	Batch	--	32.6 ± 6.0	31.7 ± 1.3	+ 3
			BLVWS	2.17 ± .17	30.8 ± 2.4		- 3
"	"	40m	Batch	--	28.3 ± 4.4	28.9 ± .9	- 2
			BLVWS	3.71 ± .5	29.5 ± 4.5		± 2
"	STA B-15	Surf	Batch	--	61.3 ± 22.4	37.5 ±20	+63
			BLVWS	1.6 ± .2	23.5 ± 1.4		-37
			BLVWS	1.9 ± .2	27.8 ± 1.4		-25
"	"	17m	Batch	--	36.2 ± 4.7	34.5 ± 3	+ 5
			BLVWS	1.7 ± .2	32.7 ± 3		- 5
"	"	37m	Batch	--	44.1 ± 9.3	41.3 ± 4	+ 7
			BLVWS	2.3 ± .2	38.4 ± 4.3		- 7
"	STA B-25	Surf	Batch	--	76.7 ± 9.7	59.7 ±24	+28
			BLVWS	2.17 ± .14	42.8 ± 5.7		-28
"	STA B-32	Surf	Batch	--	40.6 ± 9.4	32.9 ± 7	+18
			BLVWS	6.6 ± .4	28.2 ± 2		-14
			BLVWS	6.1 ± .5	29.9 ± 1		- 9
"	"	17m	Batch	--	45.6 ± 5	40.2 ± 8	+13
			BLVWS	5.0 ± .6	34.7 ± 3		-13
"	"	33m	Batch	--	44.6 ± 6	43.5 ± 1.6	+ 3
			BLVWS	10.2 ±1.6	42.4 ± 3		- 3

(continued)

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[a] Samples by LLL and PRNC were collected by the "Batch" method at a time which was usually before the long time BLVWS collections

[b] The LRE and BNWL samples were collected continuously over a time period at 2-4 hours using the BLVWS sampler which separated the particulate and soluble fractions; in 1972 two sorption beds of Al_2O_3 were used and in 1976 four Al_2O_3 beds were used.

[c] The LRE "Batch" collections were made during the BLVWS pumping to compare directly the two methods. The plutonium method of Wong et al. (1976) was employed.

The disintegration rate of the ^{232}U spike was determined by comparison of the activities of aliquots (in quadruplicate) of the ^{232}U spike and a ^{238}U standard solution electroplated simulatneously onto platinum discs. The ^{238}U solutions used for the standard were prepared by dissolving precisely weighted amounts of $99^{+}\%$ pure ^{238}U "D-38" metal supplied by the LLL.

The ^{208}Po spike was supplied as a radiochemical standard solution by the Amersham/Searle Corporation and has been calibrated several times between 1970-1975 by intercomparing the rdioactivity of plated samples with National Bureau of Standards (NBS), Battelle Northwest Laboratory (BNWL), and the LLL.

Replicate determinations of the plutonium concentration in a dissolved sediment (section 8-10 cm of core B-2) from Bikini Lagoon were performed to provide an estimate of the analytical precision of the radiochemical procedures used for plutonium analysis. The quantity of sediment (dry wt.) in each aliquote processed was 3.19 g. The chemical yield calculated from the counting data for these samples ranged from 22.6 to 40.8%. The precision for the $^{239,240}Pu$ determination was 5.3% of the mean concentration of 2. S.D. for the six analyses. The precision for ^{238}Pu measurement was 11% of the mean at 2. S.D. for the six analyses. The higher deviation about the mean for ^{239}Pu replicates is probably due to poorer counting statistics (average of 124 counts/800 minutes in the 238 peak vs. 5000 counts/800 minutes in the $^{239,240}Pu$ peak); all six ^{238}Pu concentrations found were within 2. S.D. counting errors of each other (Marshall 1975).

Quality control: problems of sample contamination were addressed by the inclusion of spiked reagent blanks with groups of samples. From several such reagent blanks, no significant contamination problem was detected. An evaluation was made of the interferences which might occur from natural and bomb-produced, alpha-emitting radionuclides in the Bikini Atoll samples.

In the plutonium and uranium procedures radium is removed along with the calcium in the chemical separation process. Isotopes of radon which might interfere are short-lived and, being gases, present no problems. Decontamination factors of greater than 1000 are reported by Butler (1968) for the removal of americium, thorium and neptunium from the final uranium samples, and similarly high decontamination factors are reported for the removal of curium and californium (Butler 1965), using TIOA separation procedures. Although Berkelium is unusual among the transamericium actinides, in that it can exist in the 4+ oxidation state (and therefore may not be separated from plutonium and uranium), it can not exist in the 4+ state in the 8 $\underline{M}$ HNO_3 - H_2O_2 solution which was used to maintain the oxidation states of $\overline{Pu}$ (VI) in the initial extraction step of the TIOA procedure (Keller 1971). The TIOA ion exchange method used in these separations provided high decontamination factors for the removal of uranium from the plutonium fraction (>300:1) and for the removal of plutonium from the uranium fraction (>1000:1) (Butler 1968).

Because no information was found concerning the plating efficiency of radionuclides which would interfere in the analysis of polonium by the plutonium procedures used in this work, solutions with known quantities of ^{241}Am, ^{242}Pu, ^{232}U, ^{228}Th, ^{224}Ra and ^{208}Po were prepared and plated as previously described.

Table B-3 shows the interlaboratory comparison results of plutonium analysis. Results of the January 1976 interlaboratory comparison of ^{210}Po in solution was Environmental Protection Agency (EPA) 164.4 ± 4.5, LRE 166 ± 5.4. The chemical procedures have been checked by the comparisons between duplicate standard samples re: IAEA, NBS, EPA. Interlaboratory comparisons between actual samples which have been exchanged are given for the results of the McClelland Laboratory (MCL) and LRE data. Of the 17 biota samples which were measured as "duplicates" six results fell outside the estimated errors of the two laboratories. It is not clear as to which laboratory was correct or whether both laboratories were correct and inhomogeniety existed in the samples. Of the five soil samples analyzed in 1971, one value was clearly outside the estimated errors of the two laboratories; and one value had a large measurement error (Nervic and Ray 1973).

A comparison of actual water samples collected in 1972 by Puerto Rico Nuclear Center (PRNC), LLL, and LRE using difference collection and analysis methods is also shown in Table B-3. Discrete samples were collected at a single time (5-10 min) by LLL and PRNC, while LRE collected samples by continuous filtration over a time period of 2-4 hours. Noshkin (1974) has shown at Enewetak that variations in ^{239,240}Pu concentrations as great as a factor of 3 can exist at certain locations over one tidal cycle.

The samples measured at Bikini, where large changes in the concentrations of Pu at different locations have been observed, compared reasonably well between the three laboratories. Values are certainly within a factor of 2 at the concentration level of pCi/1000ℓ. In fact most of the values are within 30%. Comparisons can also be made between the values of the particulate fraction of the total measured by both LLL and LRE shown in Table B-3. Most of these values are within the reported counting errors.

The direct comparison of the Batch and BLVWS methods are shown by the internal LRE intercomparisons in Table B-3. The Batch method used in these comparisons was by Wong et al. (1976); the BLVWS method employed four sorption beds of Al_2O_3 whereas only two beds were used in 1972 at Bikini and Enewetak. The Batch method and BLVWS methods compared well (average about 13% difference) on most samples with the Batch method giving slightly higher values than the BLVWS method.

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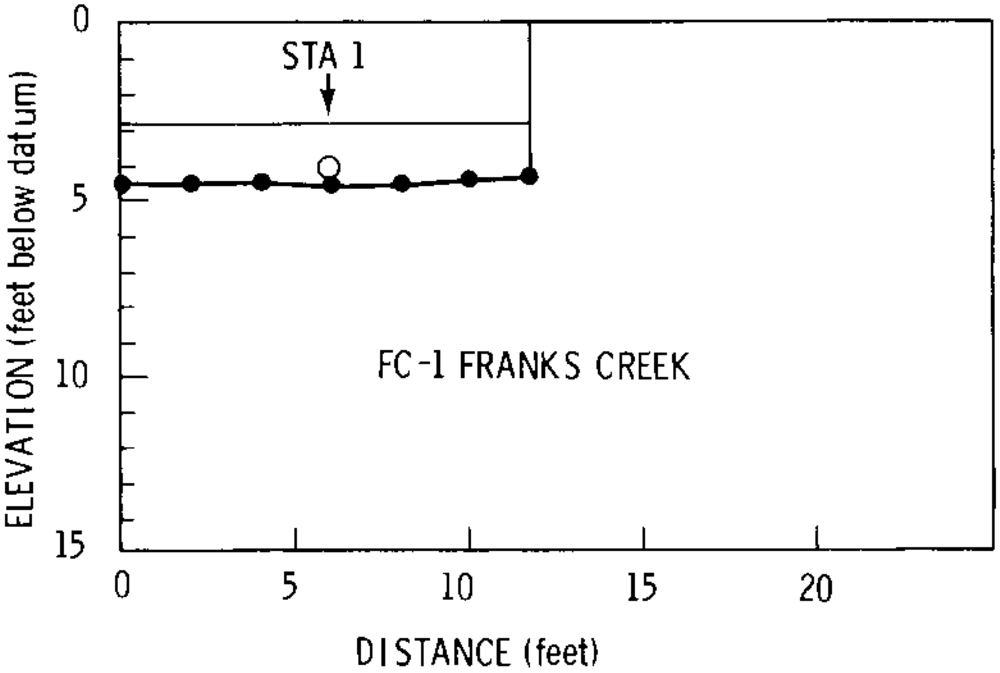
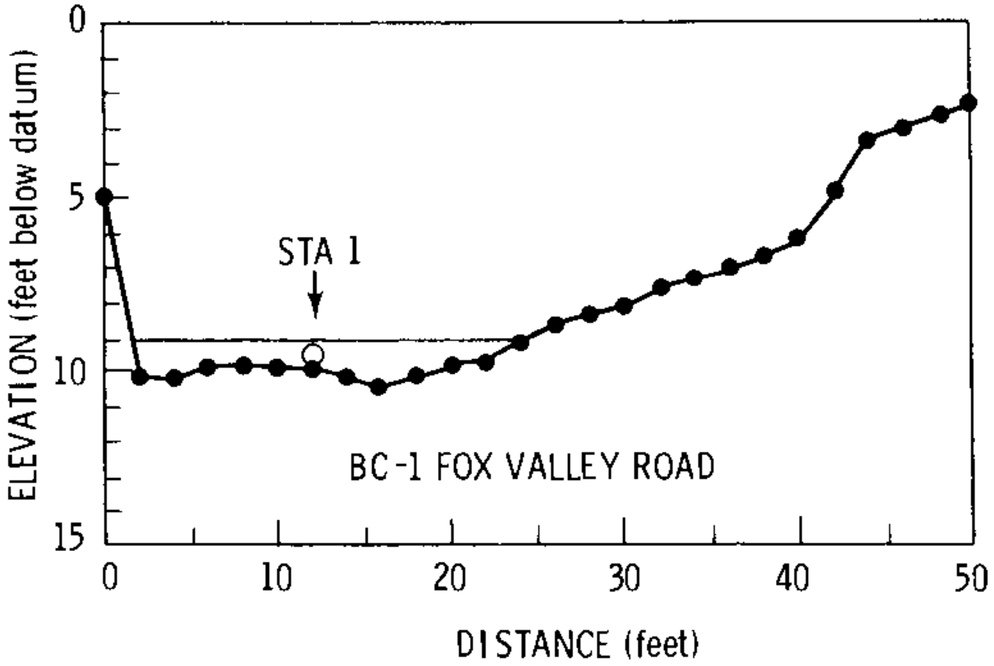
APPENDIX C

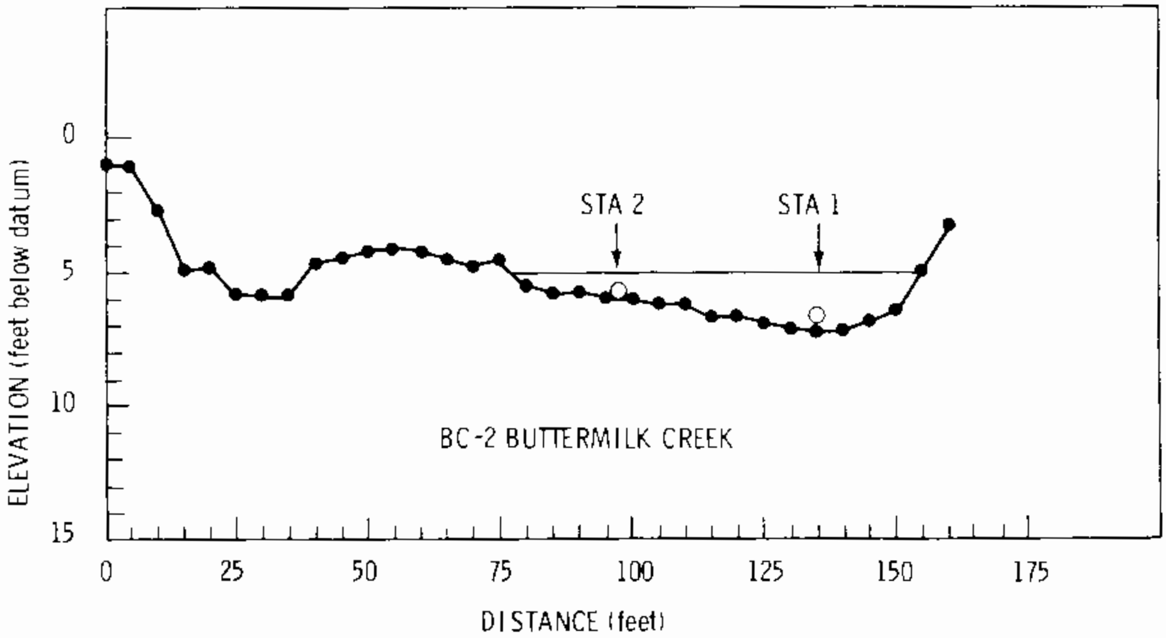
CROSS-SECTION SHAPES AND SAMPLING LOCATIONS

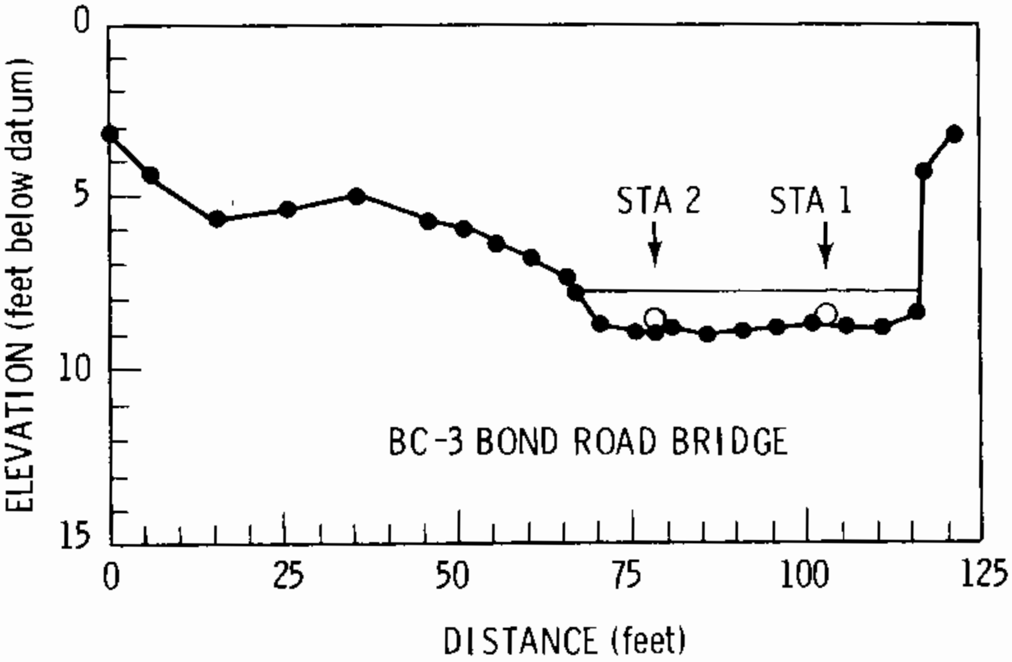


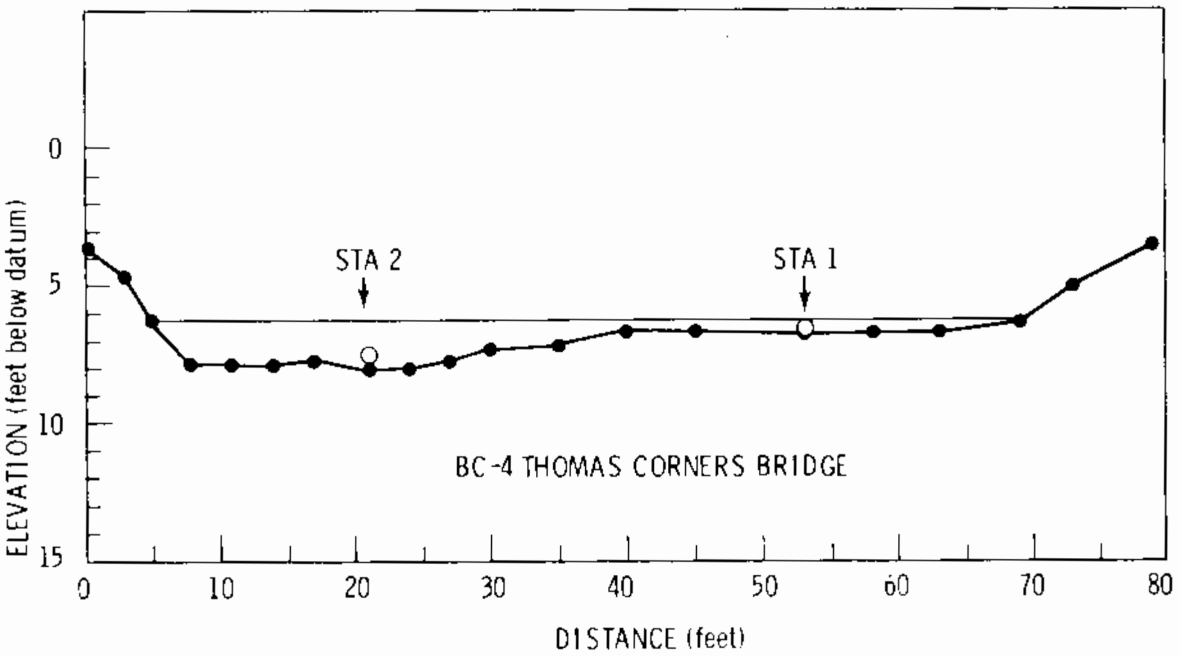
STA 1
FC-1 FRANKS CREEK
ELEVATION (feet below datum)
0
5
10
15
DISTANCE (feet)
0
5
10
15
20



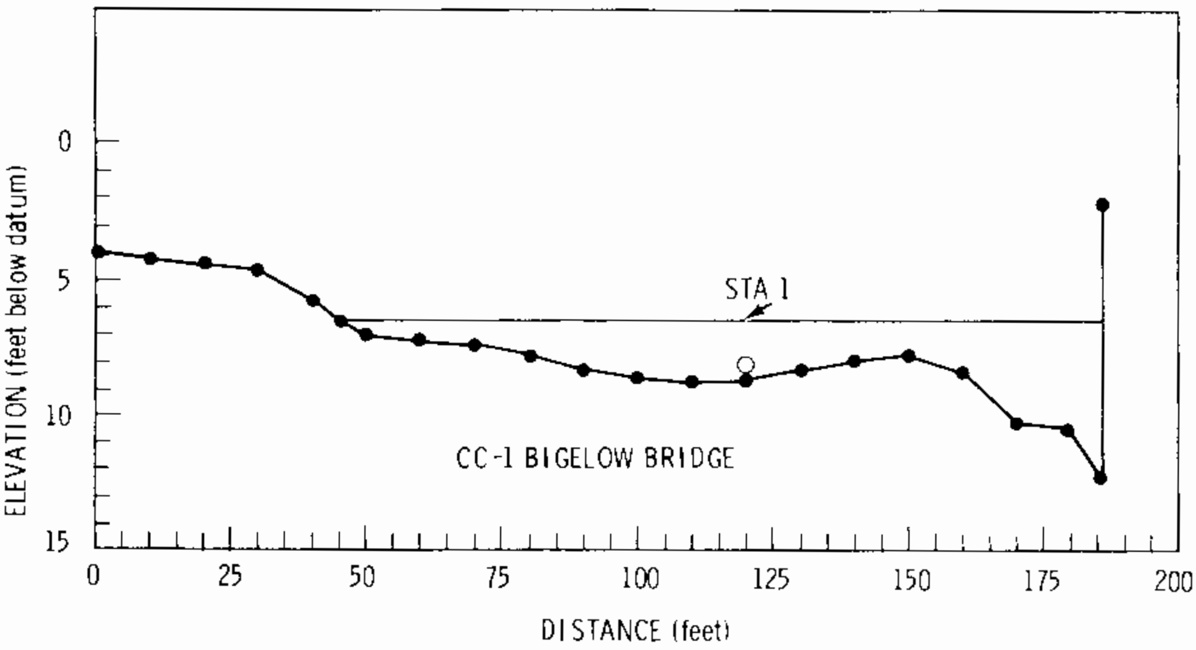
STA 1
BC-1 FOX VALLEY ROAD
ELEVATION (feet below datum)
0
5
10
15
DISTANCE (feet)
0
10
20
30
40
50


STA 2
STA 1
BC-2 BUTTERMILK CREEK
ELEVATION (feet below datum)
0
5
10
15
0
25
50
75
100
125
150
175
DISTANCE (feet)

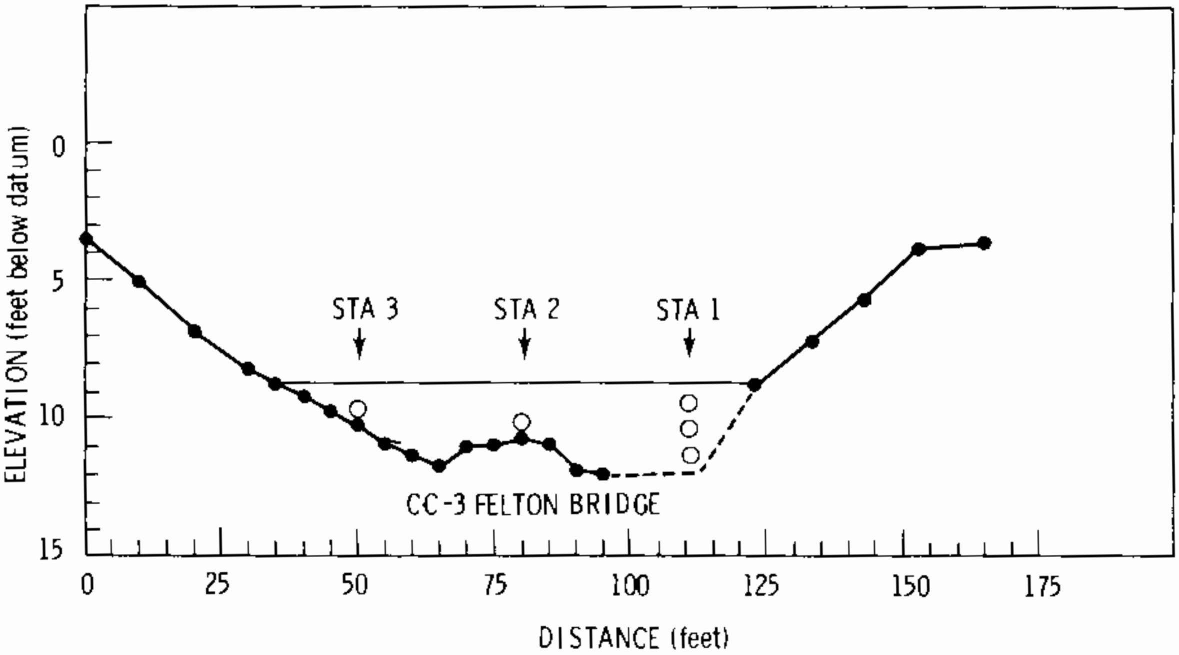

STA 2
STA 1
BC-3 BOND ROAD BRIDGE
ELEVATION (feet below datum)
0
5
10
15
0
25
50
75
100
125
DISTANCE (feet)

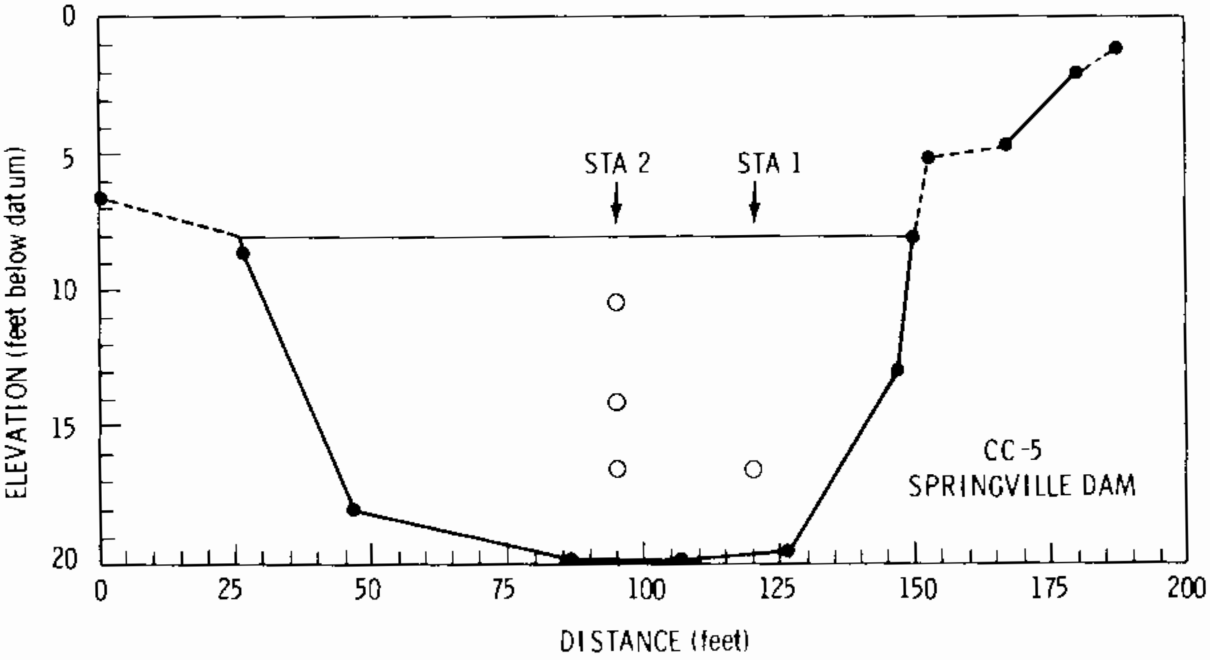


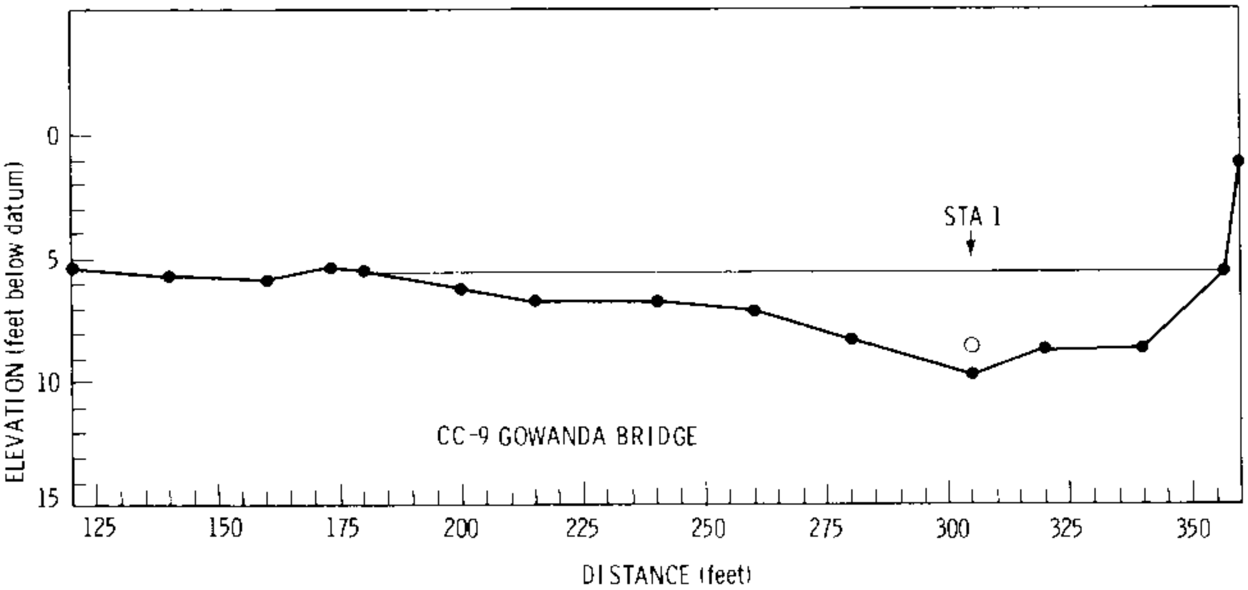
ELEVATION (feet below datum)
0
5
10
15
STA 2
STA 1
BC-4 THOMAS CORNERS BRIDGE
0
10
20
30
40
50
60
70
80
DISTANCE (feet)



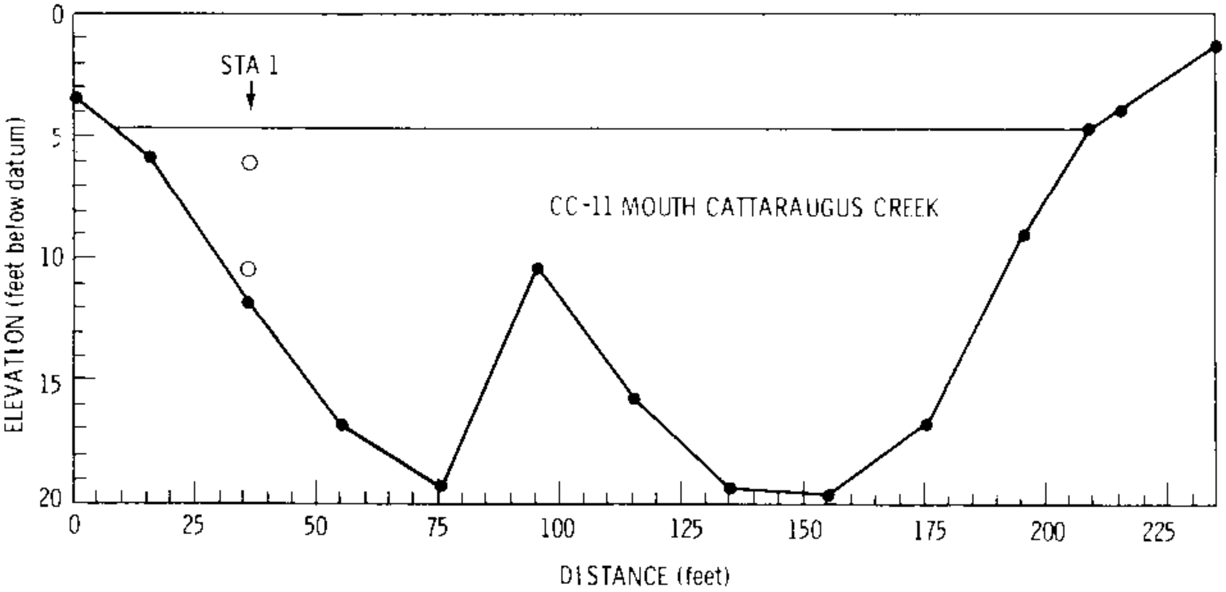
ELEVATION (feet below datum)
0
5
10
15
STA 1
CC-1 BIGELOW BRIDGE
0
25
50
75
100
125
150
175
200
DISTANCE (feet)


ELEVATION (feet below datum)
0
5
10
15
STA 3
STA 2
STA 1
CC-3 FELTON BRIDGE
0
25
50
75
100
125
150
175
DISTANCE (feet)


ELEVATION (feet below datum)
0
5
10
15
20
STA 2
STA 1
CC-5
SPRINGVILLE DAM
0
25
50
75
100
125
150
175
200
DISTANCE (feet)



ELEVATION (feet below datum)
DISTANCE (feet)
0
5
10
15
125
150
175
200
225
250
275
300
325
350
STA 1
CC-9 GOWANDA BRIDGE



ELEVATION (feet below datum)
DISTANCE (feet)
0
5
10
15
20
0
25
50
75
100
125
150
175
200
225
STA 1
CC-11 MOUTH CATTARAUGUS CREEK

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