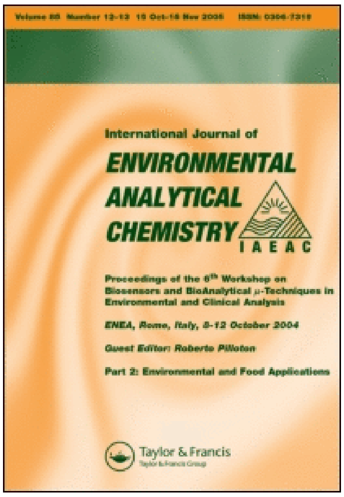

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Radioactivity from a Nuclear Fuel Reprocessing Plant Found in Natural Waters

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Radioactivity from a Nuclear Fuel Reprocessing Plant Found in Natural Waters

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Levels of tritium resulting from a commercial nuclear fuel reprocessing plant were determined to be present in Cattaraugus Creek, a stream in western New York State. The tritium levels ranged from zero to 201.4 pCi/ml in Cattaraugus Creek and up to 3889.8 pCi/ml in Buttermilk Creek, a tributary stream into which the plant effluent runs. Other radioactive species were observed in Buttermilk Creek.

KEY WORDS: Tritium, nuclear fuel plant, river water.

The world's first commercial fuels reprocessing plant is located in the southwestern portion of New York State, near West Valley, and it began operations in April 1966. This multipurpose plant is capable of processing any type of nuclear fuel elements that can be reduced to a nitric acid solution, and uses the Purex solvent extraction procedure. In April 1971, the plant completed new waste water treatment facilities, which were designed to lower the strontium and cesium content of the waters discharged into Cattaraugus Creek.

A study made in 1966, by Sax *et al.*[1], indicated that some fission products as well as tritium were being released into the environment around the plant. To determine the current levels of radioactivity in the discharge to Cattaraugus Creek, a stream that flows into Lake Erie, and the amount of tritrated water actually entering Lake Erie from this creek, a study was made at several different sampling sites located along Cattaraugus Creek.

A map showing a portion of Cattaraugus Creek and the sampling sites is given in Figure 1. The nuclear fuels reprocessing plant, which is located on Buttermilk Creek, is shown at point 1. The first sampling site, Bigelow Bridge, serves as an indicator of the tritium level in Cattaraugus Creek before the water is mixed with that of Buttermilk Creek. The next two sites, Gooseneck Creek, a tributary of Buttermilk Creek, and Buttermilk Creek are stations where the tritium levels of the water of Buttermilk Creek can be monitored before it reaches the plant. Felton Bridge is located on Cattaraugus Creek downstream from the plant, directly below the confluence of Buttermilk and Cattaraugus Creeks. Our sites on Cattaraugus Creek located in Zoar Valley and Gowanda serve as indicators of the rate of dilution of the radioactivity in the water. Another site, not shown in Figure 1, is located at the mouth of Cattaraugus Creek at Lake Erie in Silver Creek, N.Y. An

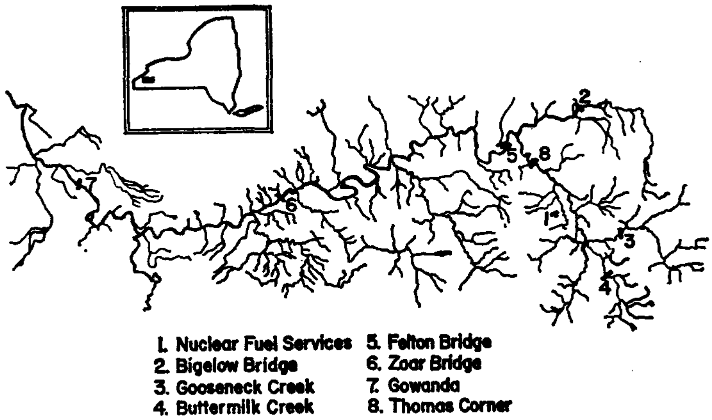

FIGURE 1 Map of Cattaraugus Creek and tributaries showing sampling stations. Insert shows position of main map in relation to the rest of New York State.

additional site, Thomas Corner Road, was added during the course of the sampling. This site is located on Buttermilk Creek between the nuclear fuels services plant and Cattaraugus Creek and serves as a more direct indicator of the levels of discharge from the plant. Samples were collected in acid-washed polyethylene containers every two weeks during the period of May 9, 1971 to October 31, 1971.

For high efficiency, low background counting, the samples were counted using liquid scintillation spectroscopy. The samples were prepared by mixing 3 ml of the water and 15 ml of Aquasol (xylene-based liquid scintillation counting solution available from New England Nuclear Corporation). The counting efficiency for tritium of this mixture was determined as $25.7 \pm 0.7\%$ by counting 10 mcl of tritiated water of known radioactivity added to the usual counting solution using distilled water. Samples were prepared in duplicate and counted for 200 min in a Packard Model 3380 Liquid Scintillation Counter.

The amount of background interference due to electronic noises, natural radioactivity, quenching, and phosphorescence in the counting vial is determined by counting 3 ml of distilled water and 15 ml of Aquasol. Plastic scintillation vials are used to reduce the interference from ^{40}K to a minimum.

The model 3380 counter is a three-channel system, and for this study the first channel was set to measure β emitters of energy between 0 to 18 keV, the second channel those of energy between 0 to 160 keV, and the third channel was used to measure high-energy beta emitters: from 160 keV to 2 MeV. With the channels set in that fashion, it is then possible to measure ^{3}H, ^{14}C, and high-energy beta emitters. The counts in the first two channels for most samples were found to be approx. equal. In order to determine whether the activity was a low or high-energy emitter, the single-channel analyzers were reset to limit the contribution of one channel to the other. Using the standard double-label setting on the scintillation system, the contribution of the ^{3}H channel to the ^{14}C channel is eliminated. The contribution of the ^{14}C channel to the ^{3}H channel is reduced so that the number of counts from a ^{14}C source in the ^{3}H channel does not exceed 10% of the counts in the ^{14}C channel. Thus, if a ^{14}C were present the number of counts in the first channel would decrease. This adjustment reduced the counts in the second channel to background levels, and it was concluded that the activity was due only to ^{3}H. Initially no counts above background were observed in the third channel.

The measured tritium levels in Cattaraugus Creek are shown in Table I. The average concentrations of tritium during the months of June through October at five of the sampling sites are shown graphically in Figure 2. The highest levels are at the Felton Bridge site which is the site closest to the nuclear fuels plant. The water reaching Silver Creek has substantially decreased tritium levels. Figure 2 also indicates the seasonal variations in the tritium levels. The amounts of tritium increase from June to July to August, where it reaches a maximum, and then decreases from September to October. These trends represent the transition from spring to summer to fall which is accompanied by increasing temperatures and decreasing amounts of rainfall through August, and then decreasing temperatures and increasing amounts of rainfall.

The influence of the amount of rainfall on the tritium levels is illustrated in Figure 3, which is a graphic representation of the recorded tritium levels at three of the sites from May 9, 1971 until October 31, 1971. The rainfall was measured in Fredonia, New York by the Vineyard Research Laboratory, New York State Agricultural Extension Service at Fredonia. From June 20 to

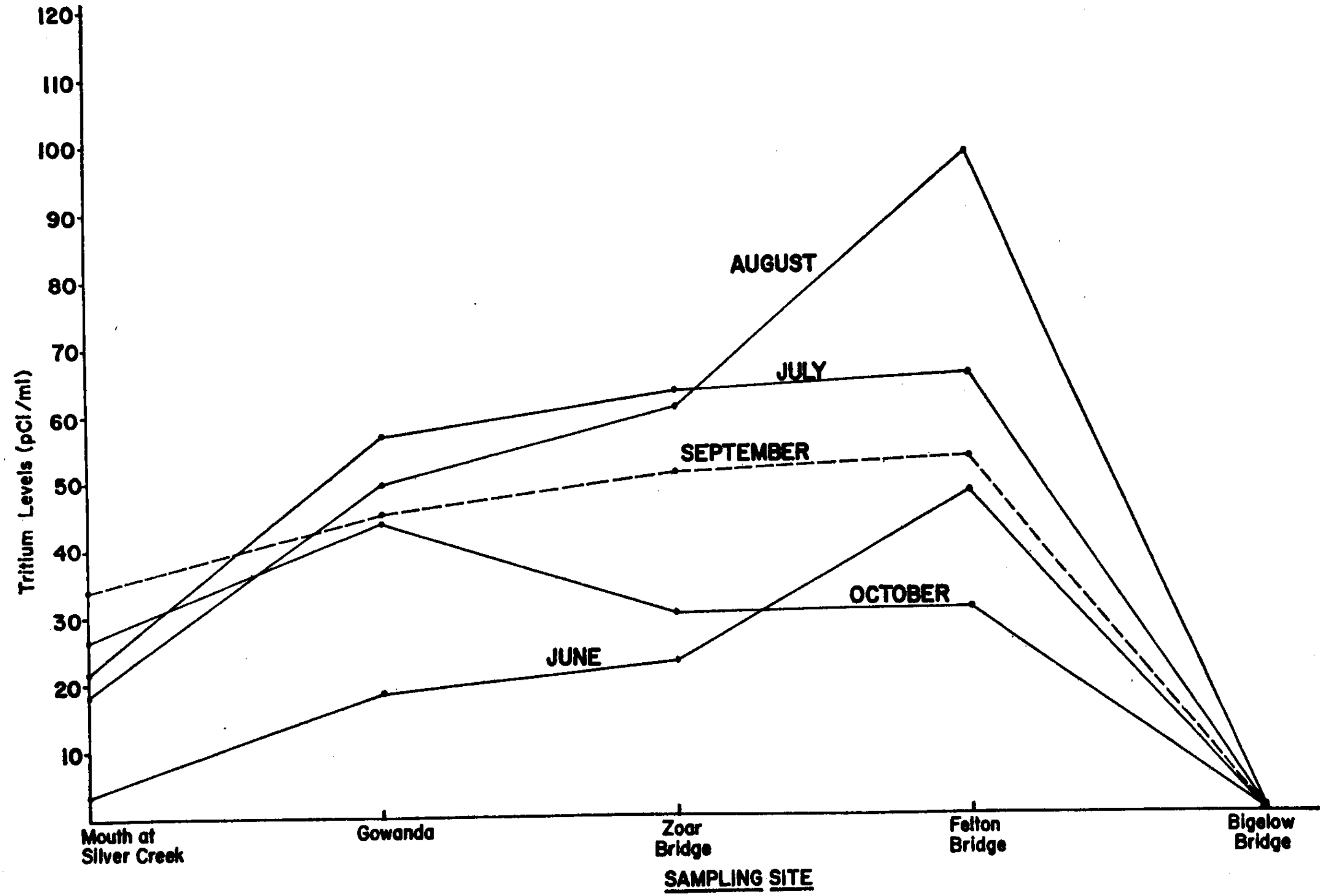

FIGURE 2 Monthly variations in the tritium levels in Cattaraugus Creek.

TABLE I

Tritium levels (pCi/ml) in Cattaraugus Creek

Sample date	Bigelow Bridge (2)[a]	Gooseneck Creek (3)	Buttermilk Creek (4)	Thomas Corner Road (8)	Felton Bridge (5)	Zoar Valley (6)	Gowanda (7)	Mouth at Silver Creek
5/9/71		0.38 ± .21			1.32 ± .21	0.78 ± .21	0.66 ± .21	0.51 ± .20
5/23/71	0 ± .21	0 ± .21	0 ± .209		2.46 ± .22	1.89 ± .22	1.47 ± .22	
6/6/71	0.80 ± .21	0.80 ± .21	0.76 ± .21		7.94 ± .23	9.57 ± .23	6.65 ± .22	3.73 ± .21
6/20/71	0 ± .15	0 ± .15	0 ± .15		87.82 ± .39	36.46 ± .28	30.29 ± .28	
7/4/71	0 ± .26	0 ± .26	0 ± .26		84.26 ± .44	71.18 ± .41	58.46 ± .39	13.69 ± .29
7/18/71	0 ± .20	0 ± .20	0 ± .20		47.22 ± .33	55.76 ± .35	55.06 ± .35	30.23 ± .29
8/1/71	0 ± .18	0.11 ± .18	0 ± .19		42.69 ± .31	33.49 ± .29	24.34 ± .26	15.70 ± .24
8/15/71	0 ± .18	0 ± .18	0 ± .18		102.51 ± .43	54.52 ± .43	41.77 ± .31	26.81 ± 27
8/19/71		10.64 ± .22		3889.77 ± 9.23	201.43 ± .57	110.57 ± .44	93.55 ± .41	
8/29/71	0 ± .18	0.16 ± .18	0 ± .18	581.34 ± 1.38	49.27 ± .33	41.99 ± .31	40.25 ± .30	12.69 ± .23
9/12/71	0 ± .16	0 ± .17	0 ± .16	1152.77 ± 1.31	79.18 ± .38	68.19 ± .34	51.20 ± .33	41.17 ± .30
9/26/71	0 ± .16	0.04 ± .16	0 ± .16	559.03 ± .92	27.93 ± .26	34.37 ± .28	39.56 ± .29	27.01 ± .26
10/10/71	0 ± .16	0 ± .16	0 ± .16	194.14 ± .56	20.19 ± .24	30.44 ± .27	33.43 ± .28	26.40 ± .26
10/31/71	0.02 ± .15	0.10 ± .15	0.10 ± .15	948.26 ± 21	41.47 ± .21	29.92 ± .27	54.33 ± .33	

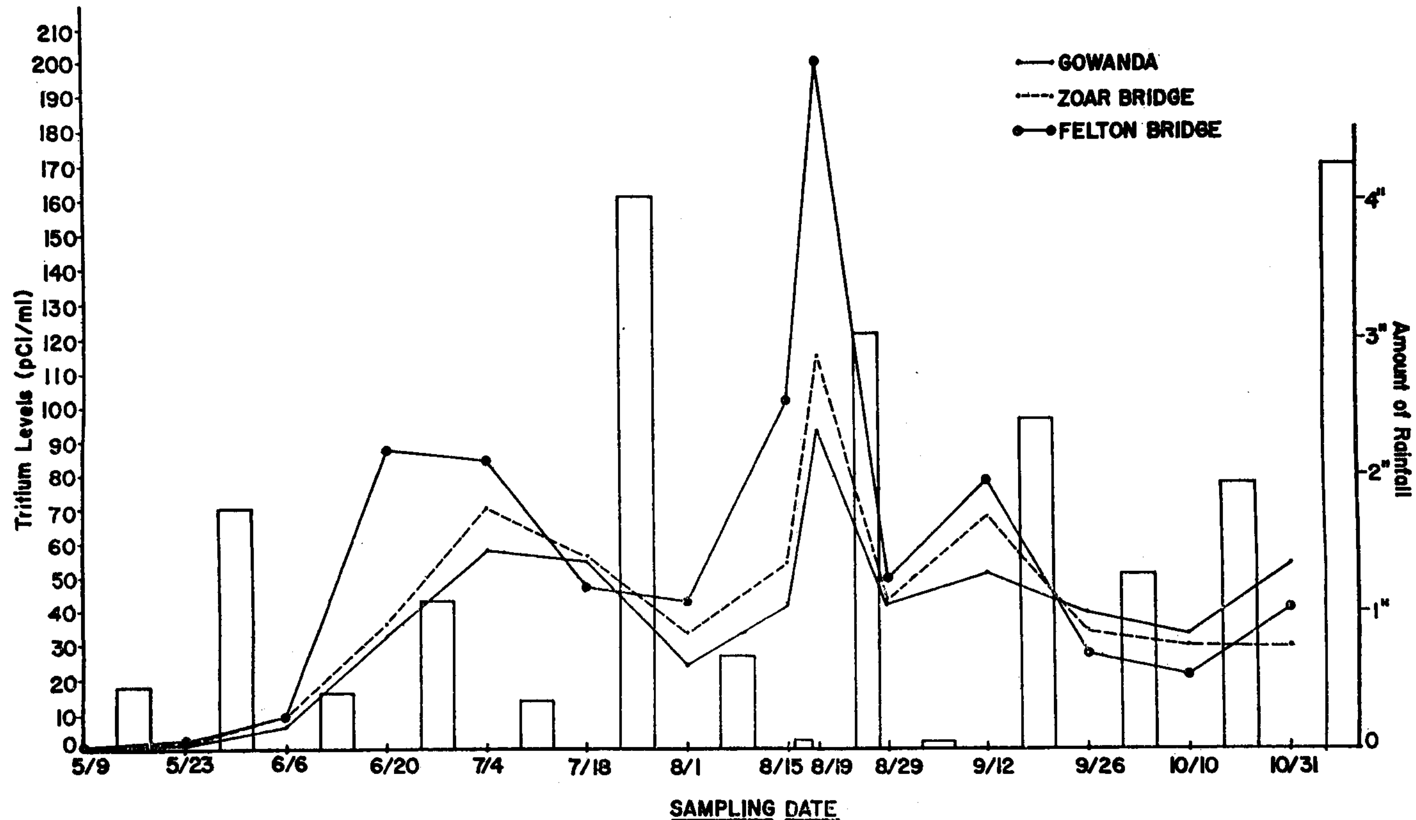

FIGURE 3 Tritium levels and rainfall plotted over the sampling period for three downstream sampling stations.

July 18, the total rainfall was only 1.52 in. and the levels are quite high. By contrast, between July 18 and July 31 the total rainfall was 4.10 in.; the levels have decreased considerably. There was no recorded precipitation between August 15 and August 19, and it was at this time that the highest tritium level of 201 pCi/ml was recorded at the Felton Bridge site.

The stream flow in Cattaraugus Creek is monitored at Gowanda, New York, by the United States Geological Survey (USGS). The stream sampling site in Gowanda is about one-half mile upstream from the stream gauge.

TABLE II

Stream flow and tritium flow at Gowanda

Date	Stream flow[a] (cubic feet/ sec)	^3H Concentration (pCi/ml	^3H flow (μCi/sec)
5/9/71	1940	0.66	36.0
5/23/71	304	1.47	12.6
6/6/71	250	6.65	57.3
6/20/71	210	30.29	180.0
7/4/71	256	58.46	424.0
7/18/71	142	55.06	221.0
8/1/71	158	24.34	109.0
8/15/71	115	41.77	136.0
8/19/71	106	93.55	281.0
8/29/71	168	40.25	192.0
9/12/71	104	51.20	151.0
9/26/71	112	39.56	125.0
10/10/71	108	33.43	102.0
10/31/71	102	54.33	157.0

[a]USGS, private communication

There are no tributaries entering the creek in this interval. Table II gives the USGS stream flow measurements and the calculated rate of tritium flow at Gowanda for the sampling dates. Linear correlation coefficients were calculated to assess the possibility of cause and effect relationships. The amount of rainfall and the ^3H concentration at the Gowanda sampling site gave an R value of -0.266. The stream flow and ^3H concentration correlation yield a coefficient of -0.496. It would appear that the activity is not due to leaching of the burial site since then increased rain would be expected to raise the level of activity. The limited correlation between stream flow and ^3H concentration would indicate that there is not a constant rate of discharge of ^3H by the reprocessing plant. If the rate of discharge were relatively constant, the concentration should then be more closely related to the amount of water available for dilution. Thus there may be some influence on tritium concentration from the amount of water in the creek, but clearly there are other major factors affecting the tritium concentration.

At the end of August, the sampling site at Thomas Corner Road was added. The tritium levels here have been found to be much higher (Table I) than at the Felton Bridge site. In addition to the increased tritium levels in Buttermilk Creek, at the site below the plant, it was noted that counts above background levels were occurring in the high-energy beta channel. These counts indicated the possibility of fission products in the water. In order to ascertain what species were present and in what concentration, gamma-ray spectra of several large (1l) samples were taken. These spectra were taken

TABLE III

Other radioactivity in Buttermilk Creek at Thomas Corner Road

Radio-nuclide	Energy of gamma rays observed (keV)	Concentration (pCi/ml)	
		10/10/71	10/31/71
^{125}Sb	176,428,463,600,631,671	$0.031 \pm .02$	$a.26 \pm 0.03$
^{106}Ru-Rh	511,616,622,1050	0.20 ± 0.15	1.68 ± 0.25
^{137}Cs	662	0	$(1.4 \pm 0.5) \times 10^{-2}$

using a nominal 55 cm^3 Ge(Li) detector with 2.1 keV FWHM at the 1332 keV line of ^{60}Co, a special plastic sample box, and a 4096 channel multichannel analyzer. The box holds 1 l of sample which completely surrounds the horizontally oriented detector. The peaks were identified on the basis of the energy of the line and the half-lives of the possible species having gamma rays of that energy. Table III lists the energies and concentrations of the species found. An absolute efficiency curve for the detector system and water sample box was obtained using the relative efficiency curve developed from external sources and from a counting standard solution of ^{60}Co in known concentrations.

The isotopes and concentrations observed in the gamma-ray spectra are given in Table III. These species are identified by their gamma-ray energies. They may not represent all of the high-energy beta emmiters observed in the third channel of the liquid scintillation spectrometer. There are several fission products which beta-decay with very weak or no gamma emission, most notably ^{90}Sr. A report by Magno *et al.*[2] indicates a number of radioactive species found in 1969 in the same location as our Thomas Corner Road site. Since that time the plant installed a new liquid effluent treatment facility. On November 4, 1969, Magno found 0.045 pCi/ml of ^{125}Sb as compared with 0.031 and 0.28 pCi/ml found on 10/10/71 and 10/31/71, respectively. They found 0.43 pCi/ml ^{106}Rh on 11/4/69, whereas 0.43 and 1.68 pCi/ml were observed in the present work. In the current study 0 and 0.014 pCi/ml of ^{137}Cs were observed, whereas Magno found 0.015 pCi. Thus the new facilities that the plant reported having installed, have not made a substantial change in the concentration of these nuclides in Buttermilk Creek.

These measured levels can be compared to the existing federal standards[3] for released radioactivity. This comparison is made in Table IV. It is seen

TABLE IV

Comparison of measured levels of radioactivity with federal standards

Nuclide	Measured concentration range (pCi/ml)	Federal concentration (pCi/ml)
^{3}H	0–3990	3000
^{125}Sb	0.03–0.28	100
^{106}Ru-Rh	0.2–1.68	10
^{137}Cs	$0–1.4 \times 10^{-2}$	20

that none of the measured levels of activity measured in Cattaraugus Creek exceed the federal standard, although the tritium content of Buttermilk Creek, downstream from the plant, does exceed the maximum permissible concentration levels on one day. It is, however, the yearly average concentration for a given species measured at the outfall of the plant that cannot exceed these federal limits. These outfall concentrations were not measured in this study.

In conclusion, the amount and the variation of the amount of tritium in Cattaraugus Creek, have been determined, It is also indicated that some low levels of fission fragments or long-lived radioactive species are being discharged into Lake Erie. In addition, the tritium levels at Silver Creek, give some indication of the extent to which the water is mixed, and exchanged with the rest of Cattaraugus Creek and its tributaries and the extent to which water is added or lost due to evaporation and precipitation before it reaches Lake Erie.

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