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IODINE-129 IN THE ENVIRONMENT AROUND A NUCLEAR FUEL REPROCESSING PLANT

U.S. ENVIRONMENTAL PROTECTION AGENCY
Office of Radiation Programs

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U.S. ENVIRONMENTAL PROTECTION AGENCY
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FOREWORD

The Office of Radiation Programs carries out a national program designed to evaluate the exposure of man to ionizing and nonionizing radiation, and to promote development of controls necessary to protect the public health and safety and assure environmental quality.

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Readers of these reports are encouraged to inform the Office of Radiation Programs of any omissions or errors. Comments or requests for further information are also invited.

W. D. Rowe
Deputy Assistant Administrator
for Radiation Programs

PREFACE

The Office of Radiation Programs of the Environmental Protection Agency, in cooperation with the New York State Department of Environmental Conservation, Nuclear Fuel Services, Inc., and the Atomic Energy Commission, have, over the past several years, conducted studies at the Nuclear Fuel Services, the nation's first commercial nuclear fuel reprocessing plant. The overall purpose of these studies is to determine the requirements of an environmental surveillance program for fuel reprocessing plants. Specific study objectives included (a) characterization of both the gaseous and liquid waste effluents from the plant, (b) measurement of the environmental concentrations of the discharged radionuclides, and (c) delineation of the critical exposure pathways and estimation of the radiation doses to the population living near the plant. The initial results of this study were published in a series of four reports:

(1) BRH/NERHL 70-1 *An Estimate of Radiation Doses Received by Individuals Living in the Vicinity of a Nuclear Reprocessing Plant*

(2) BRH/NERHL 70-2 *Liquid Waste Effluents from a Nuclear Fuel Reprocessing Plant*

(3) BRH/NERHL 70-3 *An Investigation of Airborne Radioactive Effluent from an Operating Nuclear Fuel Reprocessing Plant*

(4) BRH/NERHL 70-4 *Calibration and Initial Field Testing* ^{85}K *Detectors for Environmental Monitoring*

The results of this initial phase of the study indicated that additional information was required to better characterize the critical dose pathways and provide more accurate information for dose estimates. The environmental distribution of discharged iodine-129 was one of the subjects requiring further information. This report presents the results of followup studies on the iodine-129 discharges from Nuclear Fuel Services and on the concentrations of this radionuclide in the environment around the plant.

Charles L. Weaver
Director
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ABSTRACT

The specific activity of iodine-129, i.e., the ratio of the activity of iodine-129 to the weight of stable iodine, was measured in the environment around the Nuclear Fuel Services' reprocessing plant, West Valley, N.Y., during the summer of 1971. These measurements showed that the discharges of iodine-129 from Nuclear Fuel Services produced specific activities as high as 0.62 μCi/g of iodine in the aqueous environment and 0.28 μCi/g of iodine in the terrestrial environment around the plant. These values are about 10^4 times greater than background levels.

The average specific activity of iodine-129 in the environment around the plant was 0.1 μCi/g of iodine or 7 percent of the specific activity which if present in the human thyroid would result in the individual dose limit specified by the Federal Radiation Council.

The results of this study indicated a need for control of iodine-129 discharges from nuclear fuel reprocessing plants and surveillance programs to evaluate the environmental buildup of this radionuclide. Analytical procedures for determination of iodine-129 by neutron activation analysis and by liquid scintillation counting are included.

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IODINE-129 IN THE ENVIRONMENT AROUND A NUCLEAR FUEL REPROCESSING PLANT

INTRODUCTION

Discharges of iodine-129 to the environment from the operations of the nuclear industry are of particular interest because of the 17 million year half-life of iodine-129 and its ability to enter into the food chain and subsequently concentrate in the human thyroid. Because of its long half-life, discharged iodine-129 becomes a permanent contaminant in the environment and as a result represents a potential long-term public health problem which must be studied to determine its magnitude. Such a study, ultimately conducted by the U.S. Environmental Protection Agency, was begun in 1969 to determine the amount of iodine-129 discharged to the environment from the Nuclear Fuel Services' reprocessing plant at West Valley, N.Y., and to investigate the distribution of iodine-129 near the plant. Iodine discharge estimates for the plant's operation period of about 5 years and measurements of environmental samples taken near the plant environs during the summer of 1971 form the basis for this report.

During normal operations of nuclear power stations, negligible amounts of iodine-129 are discharged to the environment *(1)*. During subsequent chemical processing of spent fuel at a fuel reprocessing plant, a much greater potential exists for discharge to the environment of a portion of the iodine-129 present in the fuel elements. Iodine removal systems are installed in the off-gas cleaning systems at fuel reprocessing plants principally to remove iodine-131 which is still present in fuel cooled for 150 days, the current design cooling period.

The iodine removal system installed at Nuclear Fuel Services, the nation's first commercial nuclear fuel reprocessing plant, consisted of a chemical scrubber followed by silver reactors. The chemical scrubber is a 20-inch diameter by 10-foot packed column in which the dissolver off-gases are scrubbed with a stream of mercurous and mercuric nitrate solution.[1] The silver reactor, consisting of ceramic Berl Saddles coated with silver nitrate *(2)*, never achieved the design conditions and were largely ineffective for iodine absorption.[1]

Since all of the fuel processed had cooled for more than a year, no measurable iodine-131 was discharged to the environment *(3)*. However, an Environmental Protection Agency study to characterize

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[1]Personal communication from E. D. North, Director Technical Administration, Nuclear Fuel Services, Inc.

radioactive waste effluents from Nuclear Fuel Services *(4)* showed that significant quantities of iodine-129 were discharged to the environment via the stack during fuel dissolution. Data from the State of New York Department of Environmental Conservation also showed detectable concentrations of iodine-129 in deer thyroids collected around the Nuclear Fuel Services plant *(5)*. These data indicated that a detailed study of the concentrations of iodine-129 in the environment around the Nuclear Fuel Services plant was desirable to determine the effects of the iodine-129 discharges on the environment.

Russell and Hahn *(1)*, in an evaluation of the public health significance of iodine-129 from the nuclear power industry, suggested that one of the ways to evaluate the health significance of iodine-129 in the environment is to determine the specific activity of iodine-129, i.e., the ratio of activity of iodine-129 to the weight of stable iodine. This value is then compared to a limiting specific activity. For evaluation of exposures to the general population, Russell and Hahn *(1)* suggested a limiting specific activity of 1.4 μCi/g iodine as that specific activity of iodine-129 which, if present in a human thyroid, would result in a dose rate of 500 mrem/yr., the recommended Federal Radiation Council (FRC) dose limit for an individual in an exposed population *(6)*.

To evaluate the significance of the iodine-129 discharges from Nuclear Fuel Services, the Environmental Protection Agency carried out measurements of the iodine-129 and stable iodine (iodine-127) concentrations in the environment around the facility. The objectives of this study were:

(a) to measure the specific activity of iodine-129 in the environment resulting from both gaseous and liquid discharges,
(b) to compare these specific activities with the limiting specific activity as suggested by Russell and Hahn *(1)*, and
(c) to compare the specific activities around the plant with background specific activities to determine the extent of the change due to plant operations.

The environmental concentrations of iodine-129 which are presented in this report resulted from operations of the Nuclear Fuel Services plant when the iodine removal system consisted essentially of only a chemical scrubber. Nuclear Fuel Services is installing an improved iodine removal system consisting of a caustic scrubber followed by a highly efficient silver zeolite absorber.[1] Similar systems will be installed at the Midwest Fuel Recovery Plant and the Barnwell Nuclear Fuel Plant. These systems are expected to have a much greater iodine removal efficiency than the system first installed at Nuclear Fuel Services.

SAMPLE COLLECTION

Figure 1 shows the locations where samples were collected for this study; table 1 describes the sample locations and lists the distances of the locations from the plant.

Terrestrial environment

To evaluate the effect of stack discharges of iodine-129 on the environment, thyroid samples were collected from animals whose grazing habits or other limitations (fences, etc.) would restrict the intake of iodine-129 to the airborne pathway. Bovine thyroids[2] are a useful sample for this purpose since these animals graze in known restricted areas. Thyroids from woodchucks and rabbits are also useful since these animals usually graze within limited areas.[3] Animals such as deer whose grazing habits and range include: (a) the area within the high exclusion fence and (b) the stream system into which the liquid waste is discharged were not used in this study. Data from the State of New York Department of Environmental Conservation *(5)* had shown concentrations of iodine-129 in deer thyroids ranging up to 4×10^3 pCi/g of tissue. However, it was difficult to evaluate these data since the pathway through which the animal obtained the iodine-129 could not be established.

During the summer of 1971 the following samples were obtained for this study:

(1) Thyroids from woodchucks or rabbits collected at sample locations 12, 13, 14, 15, and 17.
(2) Bovine thyroids collected at a slaughterhouse from animals which had grazed at sample locations 16, 17, 19, and 20.
(3) Bovine tissues (mistakenly submitted as thyroids) collected at a slaughterhouse from animals which had grazed at sample locations 11, 18, and 21.

Aqueous environment

To evaluate the effect of liquid discharges, samples of algae and fish were collected during June 1971 from the Buttermilk and Cattaraugus Creeks, the aqueous environment into which the liquid waste

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[2]Although milk represents the pathway to man, bovine thyroids were used for this study because the thyroid is easier to analyze and permits a more sensitive measurement of the specific activity of iodine-129. For example, a 10-gram sample of bovine thyroid contained 5-10 mg of stable iodine whereas a 1-liter sample of milk would usually contain <0.1 mg of stable iodine.

[3]Personal communication from C. F. Cisar, Twinbrook Research Laboratory, Environmental Protection Agency (1972).

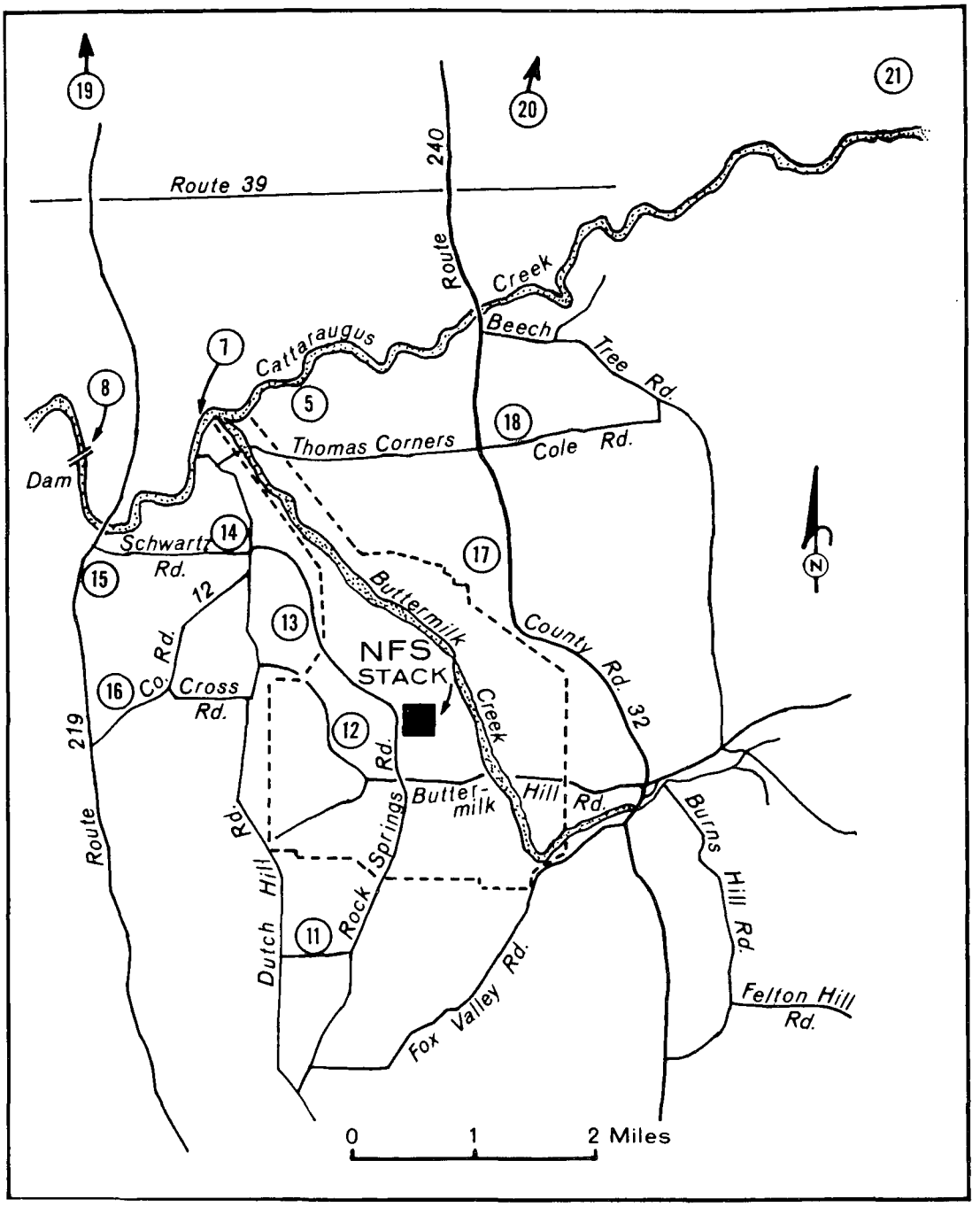

Figure 1. Sampling locations near Nuclear Fuel Services

Table 1. Location of sample collection sites

Location	Collection site number	Distance from plant stack (miles)
Rock Springs Road, Ashford	11	1.6
Rock Springs Road, Ashford	12	.2
Rock Springs Road, Ashford	13	1.0
Dutch Hill Road and Schwartz Road, Ashford	14	1.8
Schwartz Road and Route 219, Ashford	15	2.8
Ashford Hollow Road, Ashford	16	2.6
County Road - 32, Ashford	17	1.2
Thomas Corners Road, Ashford	18	2.5
Adams Road, Concord	19	6.0
Pratham Road, Concord	20	5.0
Sardina	21	10.0
Bridge at Thomas Corners Road	5	[a]3.3
Felton Bridge	7	[a]4.1
Springville Dam	8	[a]6.0

[a]Stream miles from point of discharge of liquid waste.

from Nuclear Fuel Services is discharged. Samples of each media were collected from sites 5, 7, and 8. The algae was collected from rocks in the stream bed over which the water was flowing. The fish samples were collected by rod and reel and only the tissue sections containing thyroid substance were used for analysis. Fish thyroids are not well defined or easily separated but are diffused throughout certain tissue sections[4] *(7)*. Therefore, it was not possible to analyze only the thyroids, but rather the tissue sections containing the thyroids.

Background

Iodine-129 in the environment has been attributed to fallout from nuclear weapons tests. A background iodine-129 specific activity in the environment was established from the results of analyses of bovine thyroid samples collected at a slaughterhouse in Boston, Mass., during the summers of 1970 and 1971.[5] For the purpose of this report the iodine-129 specific activities measured in these samples were considered to be representative of the general environmental specific activities for iodine-129.

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[4]Personal communication from G. LaRoche, Narraganset Marine Sport Fish Laboratory (1971).

[5]Animals had grazed in New Hampshire and Vermont.

ANALYTICAL METHODOLOGY

Because of the low specific activity of iodine-129 (1.7×10^{-4} Ci/g), activation analysis is a more sensitive technique for measuring this radionuclide in environmental samples than conventional counting techniques. Edwards *(8)* and Keisch et al. *(9)* have discussed the measurement of low concentrations of iodine-129 in environmental samples by neutron activation.

All of the iodine-129 concentrations and some of the iodine-127 concentrations (except those of the bovine thyroids) presented in this report were determined using a neutron activation procedure.

In this procedure, the iodine-129 and iodine-127 were neutron-activated to iodine-130 and iodine-128 respectively and the resultant activities measured by conventional gamma spectrometry. This procedure is summarized below and described in detail in appendix I.

When the stable iodine content of a sample exceeded 1 mg (this applied only to the bovine thyroids) a volumetric method of measurement was used. This procedure is described in appendix III. When the iodine-129 content of a sample exceeds 10 pCi, a liquid scintillation counting procedure may be used. A procedure that has been tested for this purpose at this laboratory is presented in appendix II.

Summary of activation analysis procedure

The sample is solubilized by fusion with sodium hydroxide. Iodine-131 tracer is added to determine yield. The iodine is separated from the sample by a series of solvent extraction steps with carbon tetrachloride and toluene. The iodine is then irradiated in an ammonium sulfite solution for 1 hour in a thermal neutron flux of 2×10^{13} n/cm^2 per sec. For this study the pneumatic rabbit facility at the Massachusetts Institute of Technology Research Reactor was used. Following irradiation, additional chemical purification of the iodine is accomplished by extraction with carbon tetrachloride and the purified sample is counted three times by conventional gamma spectrometry. The first count at about 1 hour after irradiation is used to determine the iodine-127; the second count 6 to 12 hours after irradiation is used to determine the iodine-129; and the third count 5 days after irradiation is used to measure the iodine-131 tracer and to correct for the yield.

The activation analysis procedure was designed to meet the objectives of this study. For more sensitive measurements of iodine-129 (and more accurate measurement of the background levels of this radionuclide) longer irradiation times and more specialized counting techniques must be used.

RESULTS AND DISCUSSION

Terrestrial environment

The results of the measurements of iodine-129 and stable iodine in small animal thyroids collected at distances from 0.2 to 3 miles from the Nuclear Fuel Services stack are presented in table 2. Table 3 presents the result of the measurements made on bovine thyroids and tissues collected from animals grazing at distances 1 to 10 miles from the point of discharge. Table 4 presents the results of measurements on bovine thyroids collected from Boston, Mass., which, for the purposes of this study, serve as background control samples.

The 2-sigma counting error associated with the measurements of iodine-127 and iodine-129 were all less than 5 percent unless otherwise noted. The overall analytical error associated with these measurements have not yet been fully evaluated. Preliminary tests indicated a precision of ± 5 to 10 percent.

The highest iodine-129 specific activity measured in small animal thyroids was 2.4×10^{-1} μCi/g iodine and this occurred in a woodchuck collected 0.2 miles from the plant. The highest specific activity measured in a bovine thyroid was 2.8×10^{-1} μCi/g of iodine and this was collected from a cow which had grazed approximately 1 mile from the plant. In general the highest iodine-129 specific activities were measured in samples collected closest to the plant and the specific activity decreased with the distance from the plant. The average iodine-129 specific activities in samples collected at various distances from the plant are presented in table 5. Although only a relatively small number of samples have been analyzed and a wide spread in the data has been observed, (particularly for the samples collected close to the plant), these data support the observation that the iodine-129 specific activity decreased with distance from the plant.

The data in table 5 show that at 5 to 6 miles from the plant the iodine-129 specific activity was 100 times greater than the background level and that even at a distance of 10 miles from the plant the iodine-129 specific activity was ten times the background level.

The average iodine-129 specific activity in samples collected 0 to 3 miles from the plant was 1.0×10^{-1} μCi/g of iodine or 7 percent of the limiting specific activity discussed by Russell and Hahn *(1)*. The specific activity data from these samples cannot be used directly to evaluate the radiation exposure from this radionuclide to the population living in the vicinity of the plant, since it is not known what fraction of the stable iodine intake for this population is of local origin. However, these data do indicate (although the plant has been operating at less than 50 percent capacity for only about 5 years) that the iodine-129 specific activities in the environment around the plant

Table 2. Iodine-129 and stable iodine in small animal thyroids collected in the vicinity of Nuclear Fuel Services

Type of sample	Date collected	Collection site	Distance and direction from plant stack (miles)	$\frac{\mu g \text{ iodine}}{g \text{ tissue (wet wt.)}}$	$\frac{\mu g\ ^{129}I}{g \text{ tissue (wet wt.)}}$	$\frac{\mu g\ ^{129}I}{g \text{ iodine}}$	$\frac{\mu Ci\ ^{129}I}{g \text{ iodine}}$
Woodchuck---	4/71	12	0.2 NW	9.4×10^{2}	1.3	1.4×10^{3}	2.4×10^{-1}
Rabbit------	5/71	14	1.8 NW	5.5×10^{2}	5.6×10^{-1}	1.0×10^{3}	1.7×10^{-1}
Woodchuck---	6/71	14	1.8 NW	2.0×10^{2}	1.5×10^{-1}	7.5×10^{2}	1.3×10^{-1}
Rabbit------	4/71	17	1.2 NE	6.2×10^{2}	3.2×10^{-1}	5.2×10^{2}	9.0×10^{-2}
Woodchuck---	5/71	17	1.2 NE	5.1×10^{2}	2.4×10^{-1}	4.7×10^{2}	8.1×10^{-2}
Woodchuck---	5/71	13	1.0 NW	4.5×10^{2}	1.8×10^{-1}	4.0×10^{2}	6.9×10^{-2}
Woodchuck---	5/71	15	2.8 WNW	9.9×10^{2}	6.0×10^{-2}	6.1×10^{1}	1.1×10^{-2}
Woodchuck---	4/71	14	1.8 NW	2.9×10^{2}	1.3×10^{-2}	4.5×10^{1}	7.8×10^{-3}

Table 3. Iodine-129 and stable iodine in bovine thyroids and tissues collected in the vicinity of Nuclear Fuel Services

Type of sample	Date collected	Collection site	Distance and direction from plant stack (miles)	$\frac{\mu g \text{ iodine}}{g \text{ tissue (wet wt.)}}$	$\frac{\mu g\ ^{129}I}{g \text{ tissue (wet wt.)}}$	$\frac{\mu g\ ^{129}I}{g \text{ iodine}}$	$\frac{\mu Ci\ ^{129}I}{g \text{ iodine}}$
Thyroid-----	9/71	17	1.2 NE	6.8×10^{2}	1.1	1.6×10^{3}	2.8×10^{-1}
Thyroid-----	9/71	16	2.1 W	4.4×10^{2}	2.4×10^{-1}	5.5×10^{2}	9.5×10^{-2}
Thyroid-----	9/71	17	1.2 NE	9.3×10^{2}	3.5×10^{-2}	3.8×10^{1}	6.6×10^{-3}
Thyroid-----	9/71	19	6.0 NNW	1.1×10^{3}	3.9×10^{-2}	3.5×10^{1}	6.1×10^{-3}
Thyroid-----	9/71	19	6.0 NNW	8.5×10^{2}	2.2×10^{-2}	2.6×10^{1}	4.5×10^{-3}
Thyroid-----	9/71	19	6.0 NNW	6.9×10^{2}	2.2×10^{-2}	3.2×10^{1}	5.5×10^{-3}
Thyroid-----	9/71	20	5.0 NNE	2.1×10^{3}	2.2×10^{-2}	1.0×10^{1}	1.7×10^{-3}
Tissue------	6/70	18	2.5 NNE	1.1	[a]6.3×10^{-4}	5.7×10^{2}	9.9×10^{-2}
Tissue------	8/71	11	1.6 S	1.4	[a]1.2×10^{-4}	8.6×10^{1}	1.5×10^{-2}
Tissue------	6/70	21	10.0 NE	4.6×10^{1}	[a]1.0×10^{-4}	2.2	3.8×10^{-4}

[a] 2σ counting error equal to $\sim 0.5 \times 10^{-4}$ μg/g tissue.

Table 4. Iodine-129 and stable iodine in bovine thyroids collected in Boston, Mass.

Sample number	Date collected	$\frac{\mu g \text{ iodine}}{g \text{ tissue (wet wt.)}}$	$\frac{\mu g\ ^{129}I}{g \text{ tissue (wet wt.)}}$	$\frac{\mu g\ ^{129}I}{g \text{ iodine}}$	$\frac{\mu Ci\ ^{129}I}{g \text{ iodine}}$
1-----	10/70	2.4×10^{2}	[a]1×10^{-4}	4×10^{-1}	7×10^{-5}
2-----	10/70	3.0×10^{2}	[b]$<1 \times 10^{-4}$	$<3 \times 10^{-1}$	$<5 \times 10^{-5}$
3-----	10/70	4.6×10^{2}	[b]$<1 \times 10^{-4}$	$<2 \times 10^{-1}$	$<3 \times 10^{-5}$
4-----	8/71	4.9×10^{2}	[a]1×10^{-4}	2×10^{-1}	4×10^{-5}
5-----	8/71	8.0×10^{2}	[a]9×10^{-5}	1×10^{-1}	2×10^{-5}
6-----	8/71	5.7×10^{2}	[a]2×10^{-4}	4×10^{-1}	7×10^{-5}

[a] 2σ counting error equal to $\sim 0.5 \times 10^{-4}$ μg/g tissue.

[b] When the sample count rate did not exceed the 2σ counting error, the activity was considered to be nondetectable and a less-than value is reported.

Table 5. Average iodine-129 specific activities at various distances from Nuclear Fuel Services

Distance from plant stack	Number of samples	Specific activity in thyroids (μCi ^{129}I/g iodine)
0 to 3 miles--------	13	1.0×10^{-1}
5 to 6 miles--------	4	4.5×10^{-3}
10 miles--------	1	3.8×10^{-4}
Background----------	6	5×10^{-5}

have increased to levels many orders of magnitude greater than background levels and to a significant fraction of the limiting specific activity.

Aqueous environment

The results of the measurements made on algae and fish samples collected from the Buttermilk and Cattaraugus Creeks are presented in table 6. The iodine-129 specific activity in the samples collected from Buttermilk Creek averaged 5.3×10^{-1} μCi/g iodine and this was a factor of about 10^4 greater than the specific activity of 5×10^{-5} μCi/g iodine measured in the background samples. The iodine-129 specific activity in samples from the Cattaraugus Creek averaged 5.8×10^{-2} μCi/g iodine or a factor of about 10^3 greater than background.

The algae and fish samples from the same locations contained similar specific activities of iodine-129 indicating that an approximate state of equilibrium had been established for iodine-129 and stable iodine in the stream system. The iodine-129 specific activity in samples from Buttermilk Creek was about ten times greater than that of samples taken from Cattaraugus Creek. This factor of ten is about equal to the dilution factor obtained when Buttermilk Creek empties into Cattaraugus Creek.

The iodine-129 specific activity in the Buttermilk Creek was 40 percent of the limiting specific activity of 1.4 μCi/g iodine as defined by Russell and Hahn *(1)*.

Background

One of the objectives of this study was to determine if the specific activity of iodine-129 in the environment around Nuclear Fuel Services was significantly different from background levels. The specific activity of iodine-129 in the Nuclear Fuel Services environment was 10^3 to 10^4 times greater than background levels of iodine-129 as measured in thyroids collected in Boston, Mass., which, for the purposes of this report, were considered to be adequate. However, since these background data were derived from only a few thyroid samples from one geographical location and since no recent data on iodine-129 in the environment are available in the literature, the limitations of the background levels of iodine-129 are recognized. Additional studies are therefore needed to establish accurately the background concentrations of iodine-129 in the environment, particularly in the environment of future nuclear fuel reprocessing plants prior to operation.

IODINE-129 DISCHARGES FROM NUCLEAR FUEL SERVICES

From April 1966 to July 1971, about 600 metric tonnes of uranium were processed by Nuclear Fuel Services with a total burnup of 4×10^6 megawatt days. The amount of iodine-129 in this fuel is estimated to have been approximately 4.4 curies *(2)*.

Table 6. Iodine-129 and stable iodine in algae and fish from the Buttermilk and Cattaraugus Creeks

Sample media	Date collected	Collection site	Location	Distance from plant[a] (miles)	μg iodine / g sample (wet wt.)	μg ^{129}I / g sample (wet wt.)	μg ^{129}I / g iodine	μCi ^{129}I / g iodine
Algae-------	6/ 9/71	5	Buttermilk	3.3	1.1	3.9×10^{-3}	3.5×10^{3}	6.1×10^{-1}
Algae-------	6/ 9/71	7	Cattaraugus	4.1	1.0	4.0×10^{-4}	4.0×10^{2}	6.9×10^{-2}
Algae-------	6/ 9/71	8	Cattaraugus	6.1	1.1	3.8×10^{-4}	3.5×10^{2}	6.1×10^{-2}
Sucker-------	6/10/71	5	Buttermilk	3.3	4.6	1.2×10^{-2}	2.6×10^{3}	4.5×10^{-1}
Trout-------	6/10/71	7	Cattaraugus	4.1	6.4	1.8×10^{-3}	2.8×10^{2}	4.8×10^{-2}
Trout-------	6/10/71	8	Cattaraugus	6.1	1.9	6.0×10^{-4}	3.2×10^{2}	5.5×10^{-2}

[a]Stream miles from point of discharge of liquid waste.

The iodine-129 discharged to the aqueous environment from April 1966 to July 1971 is estimated to have been about 0.9 curie or 20 percent of the total amount of iodine-129 in the fuel processed. This estimate is based principally on iodine-129 discharge data[6] reported by Nuclear Fuel Services *(3)*. A new low-level waste treatment facility became operational in the summer of 1971. This treatment facility is not expected to reduce the iodine-129 discharges significantly unless additions are made to the facility for this purpose.

No data were available on stack discharges of iodine-129 for the period April 1966 to January 1971. However, for the purposes of this report, the stack discharges of iodine-129 for this period were roughly estimated by the following method:

An activated charcoal trap used by Nuclear Fuel Services to sample iodine stack discharges for the period April through June 1971 was analyzed for iodine-129 by the Environmental Protection Agency. This analysis and data based on the burnup of fuel processed during this period, indicated that 25 percent of the iodine-129 present in the fuel was discharged to the environment via the stack. This value is probably a minimum value, since the efficiency of the activated charcoal for retaining iodine is unknown.[7] If this value of at least 25 percent was representative of the iodine-129 discharged via the stack during the entire period of operation, then a minimum of 1.1 curies of iodine-129 were discharged to the environment by this route for the period April 1966 to July 1971.

SUMMARY AND CONCLUSIONS

During the first 5 years of operation of the Nuclear Fuel Services plant, discharges of iodine-129 have resulted in specific activities of this radionuclide as high as 6.1×10^{-1} μCi/g of iodine in the aqueous environment and 2.8×10^{-1} μCi/g of iodine in the terrestrial environment around the plant. These values are a factor of about 10^4 higher than background levels. Even at a distance of 10 miles from the plant, the iodine-129 specific activity was ten times the background.

The average specific activity in the terrestrial environment around the plant perimeter was 1×10^{-1} μCi/g of iodine or 7 percent of the limiting specific activity. Although the specific activity data could not be used to estimate the radiation exposure to the population living in the vicinity of the plant, it did indicate a rapid buildup of iodine-129 in the plant environs, which requires careful consideration and further evaluation.

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[6]No liquid discharge data were available for April 1966 through December 1966, and the discharge estimated for this period was based on the amount of fuel processed.

[7]Personal communication from J. H. Keller, Idaho Nuclear Corporation (1971).

Data on iodine-129 discharges from Nuclear Fuel Services indicate that a large percentage of the iodine-129 present in the spent fuel elements will be volatilized during fuel reprocessing. Most of this liberated iodine-129 will be removed by the off-gas cleaning system. For a 1,500 ton-per-year plant, as much as 800 pounds of iodine-129 per year could be retained by the off-gas cleaning system. Details are not available on the method of disposal planned for the components of the off-gas cleaning systems of fuel reprocessing plants. However, procedures which would result in permanent storage of the iodine-129 present in these components would seem to be indicated.

The results of this study indicate a need for controlling iodine-129 discharges from nuclear fuel reprocessing plants and surveillance programs[8] to evaluate the environmental buildup of this radionuclide. The recommendations presented below are directed toward these areas.

RECOMMENDATIONS

(1) To keep the stack discharge of iodine-129 as low as practicable, all nuclear fuel reprocessing plants should be required to operate their iodine removal system even when processing aged fuel in which the iodine-131 content has decayed to a small quantity.[9]
(2) Environmental surveillance programs at nuclear fuel reprocessing plants should include periodic measurements of the specific activity of iodine-129 in the plant environs.[10]
(3) Careful consideration should be given to the methods used for the disposal of the scrubber solutions and absorbers used in iodine removal systems.
(4) A detailed research study of the behavior of iodine-129 in the Nuclear Fuel Services plant environmental ecosystem should be carried out. Information should also be obtained on the stable iodine intake for the population living in the vicinity of the plant to determine what percentage of their total iodine intake is of local origin. Measurements of iodine-129 in human thyroids from individuals who had lived in the vicinity of the plant would be desirable whenever possible. As a part

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[8]Similar to those carried out by the New York State Department of Environmental Conservation.

[9]This recommendation is in contrast to those of Bryant *(10)* and Rodger *(11)* who have indicated that discharges of iodine-129 to the environment will not be a serious problem for plants processing less than 5 to 10 tonnes of fuel per day.

[10]Yearly measurements of iodine-129 and stable iodine in bovine thyroids or in milk collected around the plant should provide the required information.

of this study, background specific activities of iodine-129 should also be established, both in the general environment and in the environs of fuel reprocessing plants prior to facility operations.

ADDENDUM

Measurements of iodine-129 were made on a series of milk samples collected by the New York State Department of Environmental Conservation during the spring of 1972 from farms located in the vicinity of Nuclear Fuel Services. These measurements using activation analysis were carried out in order to verify the detection of iodine-129 in milk by the New York State Department of Environmental Conservation *(12)* whose measurements were made using the liquid scintillation technique (appendix II). The results of these measurements are presented in table 7. These data showed detectable iodine-129 in all samples collected. The iodine-129 concentrations ranged from 0.05 to 1.51 pCi/liter.

Table 7. Iodine-129 concentrations in milk collected from farms located in the vicinity of Nuclear Fuel Services

Date collected	Distance from plant stack (miles)	Iodine-129 (pCi/liter)
3/31/72--------	2.0	1.51
4/ 4/72--------	1.5	1.04
3/28/72--------	1.7	.87
4/ 3/72--------	2.0	.60
3/31/72--------	3.0	.35
3/28/72--------	5.0	.30
4/ 4/72--------	7.0	.19
3/29/72--------	5.0	.11
4/ 3/72--------	5.0	.06
3/30/72--------	7.0	.05
4/ 6/72--------	Albany, N.Y.	< .001

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APPENDIX I

PROCEDURES FOR THE ANALYSES OF IODINE-129 AND IODINE-127 IN ENVIRONMENTAL SAMPLES

The procedures described below have been specifically written for the analysis of thyroid samples. These procedures are also equally applicable to other environmental samples. However, if the sample size exceeds 10 grams, then the "Sample Preparation" and "Iodine Extraction" procedures would have to be scaled up appropriately.

When the iodine-129 content of a sample is known to exceed 5 to 10 pCi, then determination by liquid scintillation counting is recommended. For smaller amounts of iodine-129, determination by activation analysis is recommended. Likewise, when the stable iodine content of a sample exceeds 1 mg then it may be determined by titrimetry; otherwise it can be determined simultaneously with the iodine-129 by activation analysis.

SAMPLE PREPARATION

1. Cut the thyroid (5 to 10 grams) into small pieces and place in a 250-ml nickel crucible. If the sample is to be analyzed by neutron activation, add about 1,000 dpm of carrier-free iodine-131 for determination of the chemical recovery.
2. Add 10 ml of water, 30 grams of NaOH pellets and, carefully, 10 ml of ethyl alcohol.
3. Heat gently on a hot plate, stirring as needed, to solubilize the mass. Evaporate off the ethyl alcohol.
4. Place the crucible in a muffle furnace at 250° C. Raise the temperature to 600° C. in increments of 100 degrees at 15 to 30 minute intervals.
5. Remove the crucible from the muffle furnace and cool. Dissolve the melt with water and transfer to an 800-ml beaker. Wash the crucible with water.

IODINE EXTRACTION

1. Heat the solution from step 5 of the "Sample Preparation" procedure to just below boiling on a hot plate. Add additional water if necessary (volume should be about 400 ml). Slowly pass chlorine gas through the solution for about 30 seconds.
2. Cool the solution. Acidify using concentrated nitric acid (pH < 1) and transfer to a 1,000-ml separatory funnel.
3. Add 50 ml of carbon tetrachloride and 10 ml of 1 M hydroxylamine hydrochloride. Shake for 2 minutes to extract the iodine into the organic phase. Draw off the organic layer into a 250-ml separatory funnel.

4. Add 25 ml of carbon tetrachloride and 5 ml of 1 M hydroxylamine hydrochloride to the first separatory funnel and shake for 2 minutes. Combine the organic phases. Discard the aqueous phase.
5. Add 25 ml water and 10 drops of 1 M sodium sulfite to the separatory funnel containing the carbon tetrachloride. Shake for 1 to 2 minutes. Discard the organic phase.
6. Add 20 ml of toluene, 1 ml of 1 M nitric acid, and 10 drops of 1 M sodium nitrite to the separatory funnel. Shake for 1 to 2 minutes. Discard the aqueous phase.
7. Add 10 ml of water and 10 drops of 1 M sodium sulfite to the separatory funnel and shake for 1 to 2 minutes. Transfer the aqueous phase to a 60-ml separatory funnel. Discard the organic phase.
8. Add exactly 10 ml of toluene, 1 ml of 1 M nitric acid and 10 drops of sodium nitrite to the separatory funnel and shake for 2 minutes. Discard the aqueous phase.
9. Add 10 ml of 0.01 M nitric acid to the separatory funnel and shake for 30 seconds. Discard the aqueous phase.
10. Repeat step 9.
11. If the sample is known to contain more than 5 to 10 pCi of iodine-129, proceed with the "Determination of Iodine-129 by Liquid Scintillation Counting" (appendix II); otherwise proceed with the "Determination of Iodine-129 by Neutron Activation." If there is a distinct pink color to the toluene layer, use the procedure for the "Volumetric Determination of Stable Iodine" (appendix III); otherwise determine the stable iodine (iodine-127) simultaneously with the iodine-129 by neutron activation.

DETERMINATION OF IODINE-129 AND IODINE-127 BY NEUTRON ACTIVATION

1. Take a 5 to 10 ml aliquot of the toluene layer from step 10 of the "Iodine Extraction" procedure and place this aliquot in a 60-ml separatory funnel.
2. Add 4 ml of 1 M ammonium sulfite to the separatory funnel. Shake for 2 minutes and transfer the aqueous phase to a 10-ml volumetric flask.
3. Repeat step 2, combining the aqueous portions in the volumetric flask and dilute to volume with 1 M ammonium sulfite. Transfer 5 ml of this solution to a polyethylene irradiation vial and heat seal.
4. Prepare standards of iodine-127 and iodine-129 as follows: Pipet exactly 5 ml of a stock solution containing a known amount of iodine-127 (10 μg/ml) in 1 M ammonium sulfite into a polyethylene vial and heat seal. Similarly, pipet exactly 5 ml of a stock solution containing a known amount of iodine-129 (0.01 μg/ml) in 1 M ammonium sulfite into another polyethylene vial and heat seal.
5. Irradiate the vials containing the samples and the standards in a constant neutron flux of about 2×10^{13} n/cm^2-sec. for 60 minutes. Record the exact time of removal from the flux. Allow the

radioactivity in the vials to decay for at least 10 minutes.[1]

6. Cut open a vial carefully and transfer the solution (pipet exactly 4 ml of the standard solutions) to a 60-ml separatory funnel containing 10 ml of water, 10 mg of iodine carrier (as potassium iodide), and 20 ml of toluene.
7. Acidify with 6 ml of 1 M nitric acid. Add 2 ml of 1 M sodium nitrite to completely oxidize the iodine to free iodine. Shake for about 2 minutes. Discard the aqueous phase.
8. Wash the organic phase three times, shaking for 30 seconds with 10 to 15 ml of 0.01 M nitric acid each time. Discard washings.
9. Add 15 ml of 1 M ammonium sulfite. Shake for 1 minute. Transfer the aqueous portion to a gamma counting container.
10. Add 15 ml of 1 M ammonium sulfite to the separatory funnel containing the toluene and shake for 1 minute. Combine the aqueous portion with the aqueous portion from step 9 and adjust the volume to a predetermined geometry for gamma counting.
11. Count the sample gamma spectrometrically three times at the following intervals:

    (a) immediately after chemical separation for the measurement of iodine-128;
    (b) 6 to 12 hours after irradiation for the measurement of iodine-130; and
    (c) 5 days after irradiation for the measurement of iodine-131 to determine chemical recovery.

CALCULATIONS

$$^{129}I \text{ or } ^{127}I \ \mu g/gram = \frac{1}{C\,E}\left(\frac{A}{D} - \frac{B}{F}\right)$$

A = Net cpm of iodine-128 or iodine-130 in the sample as determined from the gamma count data and corrected for decay to time of removal from the neutron flux (Note 1).

B = Net cpm of iodine-128 or iodine-130 as determined from the gamma count data of a reagent blank and corrected for decay to time of removal from the neutron flux.

C = Net cpm of $^{128}I/\mu g\ ^{127}I$ or net cpm of $^{130}I/\mu g\ ^{129}I$ at the time of removal from the neutron flux as determined from the gamma count data of the standards.

D = Overall chemical recovery of iodine in the sample (Note 2). This value includes the correction for the aliquot taken for irradiation.

E = Weight of the sample in grams.

F = Overall chemical recovery of iodine in the blank (Note 2). This value includes the correction for the aliquot taken for irradiation.

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[1]In practice 30 to 60 minutes elapsed between the end of the irradiation and the beginning of the postirradiation chemistry.

Note 1: The net cpm of iodine-128 and iodine-130 are determined by measuring the 0.44 MeV and 0.53 MeV photopeaks respectively using a method of simultaneous equations similar to that described by Hagee.[2] For low-count rates spectral interferences from iodine-131 and sodium-24 may also have to be considered.

Note 2: The chemical recovery is determined as follows:

$$D = \frac{\text{Net cpm } ^{131}\text{I in sample or blank}}{\text{Net cpm } ^{131}\text{I in tracer added to sample or blank}}$$

The net cpm of iodine-131 is determined from the measurement of the 0.36 MeV photopeak as described in Note 1.

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[2]Hagee, G. A., G. J. Karches and A. S. Goldin. Determination of ^{131}I, ^{137}Cs, and ^{140}Ba in Fluid Milk by Gamma Spectroscopy. Talanta 5:36 (1960).

APPENDIX II

DETERMINATION OF IODINE-129 BY LIQUID SCINTILLATION COUNTING

1. Pipet exactly 4 ml of toluene from step 10 of the "Iodine Extraction" procedure into a liquid scintillation counting vial. Add 1 ml of 2-methyl-1-butene and 10 ml of scintillation solution (Note 1).
2. Cap the vial and place it under a fluorescent light for about 2 hours to decolorize the iodine.
3. Count for iodine-129 in a liquid scintillation counter.

Note 1: The scintillation solution is prepared by dissolving 7 grams of 2,5-diphenyloxazole (PPO) and 0.1 gram of 1,4-bis-2 (5-phenyl-oxazolyl)-benzene (POPOP) in 1 liter of toluene.

CALCULATION

$$\text{pCi } ^{129}\text{I/gram of sample} = \frac{AB}{2.22\ CDE}$$

A = Net cpm of iodine-129 in aliquot of sample counted.
B = Volume (ml) of toluene added in step 8 of "Iodine Extraction" procedure (10 ml).
C = Counting efficiency of iodine-129 (cpm/dpm).
D = Volume (ml) of toluene taken for analysis (4 ml).
E = Weight of sample taken for analysis (grams).

APPENDIX III

VOLUMETRIC DETERMINATION OF STABLE IODINE

1. Pipet a 4-ml aliquot of the toluene layer from step 10 of the "Iodine Extraction" procedure, into a 60-ml separatory funnel. Add about 20 ml of water and 5 ml of 1 M sodium sulfite dropwise to completely reduce the iodine to iodide. Shake for about 2 minutes.
2. Collect the aqueous portion in a 125-ml Erlenmeyer flask.
3. Add another 20 ml of water to the organic phase, and 10 drops of sodium sulfite. Shake for 2 minutes. Combine the aqueous phase with the aqueous portion from step 2. Discard the organic phase.
4. Add about 5 ml of bromine-sodium acetate solution (Note 1) and heat to boiling to insure complete oxidation of the iodide to the iodate.
5. Cool in an ice bath. Add formic acid (88 to 90 percent) dropwise until the yellow color of the bromine disappears (Note 2).
6. Add about 3 ml of 10 percent potassium iodide solution, 10 ml of 1 M sulfuric acid, and about 5 drops of 1 percent starch solution.
7. Titrate the liberated iodine with a standardized 0.02 N sodium thiosulfate solution (Note 3). Color change is from blue to colorless at the endpoint.

Note 1: The bromine-sodium acetate reagent is prepared by dissolving 10.0 grams of sodium acetate trihydrate and 2 ml of bromine in 100 ml of glacial acetic acid.

Note 2: The solution must be cooled before the excess bromine is destroyed with formic acid. If the solution is too warm, the formic acid will also reduce some of the iodate giving low results. It is also important that the solution be colorless after the bromine is destroyed with formic acid.

Note 3: The 0.02 N thiosulfate solution is prepared as follows: Dissolve 2.5 grams of $Na_2S_2O_3 \cdot 5H_2O$, 0.2 grams Na_2CO_3, and 3 drops of chloroform in water and make up to 1 liter. Standardize by titrating against a known amount of iodine to obtain a titer in ml of thiosulfate solution per mg of iodine.

CALCULATION

$$\text{mg of iodine/gram of sample} = \frac{A\ B}{C\ D\ E}$$

A = Volume (ml) of thiosulfate solution used to titrate sample.
B = Volume of toluene added in step 8 of "Iodine Extraction" procedure (10 ml).
C = Titer: ml of thiosulfate solution per mg of iodine.
D = Volume of toluene taken for stable iodine determination (4 ml).
E = Weight of sample taken for analysis (grams).

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IODINE-129 IN THE ENVIRONMENT AROUND NUCLEAR FUEL REPROCESSING PLANT, ORP/SID 72-5. P. J. Magno, T. C. Reavey and J. D. Apitianakis (November 1972)

ABSTRACT: The specific activity of iodine-129, i.e., the ratio of the activity of iodine-129 to the weight of stable iodine, was measured in the environment around the Nuclear Fuel Services' reprocessing plant, West Valley, N.Y., during the summer of 1971. These measurements showed that the discharges of iodine-129 from Nuclear Fuel Services produced specific activities as high as 0.62 μCi/g of iodine in the aqueous environment and 0.28 μCi/g of iodine in the terrestrial environment around the plant. These values are about 10^4 times greater than background levels.

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The average specific activity of iodine-129 in the environment around the plant was 0.1 μCi/g of iodine or 7 percent of the specific activity which if present in the human thyroid would result in the individual dose limit specified by the Federal Radiation Council.

The results of this study indicated a need for control of iodine-129 discharges from nuclear fuel reprocessing plants and surveillance programs to evaluate the environmental buildup of this radionuclide. Analytical procedures for determination of iodine-129 by neutron activation analysis and by liquid scintillation counting are included.

KEY WORDS: Environment; iodine-129; New York State; nuclear fuel reprocessing plants; thyroids.

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