

IODINE-129 IN THE SNAKE RIVER PLAIN AQUIFER AT AND NEAR THE IDAHO NATIONAL ENGINEERING LABORATORY, IDAHO, 1990-91

**By Larry J. Mann, U.S. Geological Survey, and
T.M. Beasley, U.S. Department of Energy**

**U.S. GEOLOGICAL SURVEY
Water-Resources Investigations Report 94-4053**

**Prepared in cooperation with the
U.S. DEPARTMENT OF ENERGY**

**Idaho Falls, Idaho
April 1994**



U.S. DEPARTMENT OF THE INTERIOR

BRUCE BABBITT, Secretary

U.S. GEOLOGICAL SURVEY

Gordon P. Eaton, Director

Any use of trade, product, or firm names is for descriptive purposes only and does not imply endorsement by the U.S. Government.

For additional information write to:

U.S. Geological Survey
INEL, MS 4148
P.O. Box 2230
Idaho Falls, ID 83403

Copies of this report can be purchased from:

U.S. Geological Survey
Earth Science Information Center
Open-File Reports Section
Box 25286, Mail Stop 517, Denver Federal Center
Denver, CO 80225

Contents

Abstract.....	1
Introduction.....	1
Physical setting	3
Previous investigations	3
Field methods.....	4
Analytical methods and quality assurance.....	4
Guidelines for the interpretation of analytical results.....	6
Acknowledgments	7
Occurrence and characteristics of iodine-129.....	7
Concentrations of iodine-129 in the Snake River Plain aquifer	8
Areal distribution of iodine-129 in the Snake River Plain aquifer	14
Vertical distribution of iodine-129 in the Snake River Plain aquifer	21
Iodine-129 movement and ground-water flow velocities in the Snake River Plain aquifer.....	24
Comparison of iodine-129 concentrations to drinking-water standards.....	24
Summary	25
References cited	25

Illustrations

Figure 1. Map showing location of the Idaho National Engineering Laboratory.....	2
2. Graph showing mean annual concentrations of iodine-129 in wastewater discharged to the disposal well and ponds at the Idaho Chemical Processing Plant.....	15
3. Graph showing annual discharge of the Big Lost River below INEL diversion.....	16
4. Graph showing mean concentrations of iodine-129 and the experimental standard errors of the means in water from 18 wells sampled in 1977, 1981, 1986, and 1990-91, Idaho National Engineering Laboratory	17
5. Map showing areal distribution of iodine-129 in the Snake River Plain aquifer near the Idaho Chemical Processing Plant, 1990-91.....	18
6. Map showing areal distribution of iodine-129 in the Snake River Plain aquifer near the Idaho Chemical Processing Plant, 1986.....	19
7. Map showing distribution of iodine-129 in the Snake River Plain aquifer in areas adjacent to the Idaho Chemical Processing Plant, 1990-91	20
8. Graph showing depth to water in well 50 at the Idaho Chemical Processing Plant.....	23

Tables

Table 1. Depths of wells, intervals of wells open to aquifer, depths to water and dates water samples were collected for iodine-129 analyses, Idaho National Engineering Laboratory.....	9
2. Iodine-129 concentrations in ground water, Idaho National Engineering Laboratory, 1986 and 1990-91,	11

Conversion Factors and Abbreviated Units

Multiply	By	To Obtain
foot (ft)	0.3048	meter
mile (mi)	1.609	kilometer
square mile (mi ²)	2.590	square kilometer
acre-foot (acre-ft)	1,233	cubic meter
curie (Ci)	3.7×10^{10}	becquerel
picocurie per liter (pCi/L)	0.037	becquerel per liter
millirem	0.01	millisievert
dram	3.888	gram

For temperature, degrees Celsius (°C) may be converted to degrees Fahrenheit (°F) by using the formula $^{\circ}\text{F} = (1.8)(^{\circ}\text{C}) + 32$.

Abbreviations for units: L (Liter), mL (milliliter), mg/mL (milligram per milliliter), and mg/L (milligram per liter).

Iodine-129 in the Snake River Plain Aquifer at and near the Idaho National Engineering Laboratory, Idaho, 1990–91

By Larry J. Mann, U.S. Geological Survey, Idaho Falls, Idaho and
T.M. Beasley, U.S. Department of Energy, Environmental Measurements
Laboratory, New York, New York

Abstract

From 1953 to 1990, an estimated 0.56 to 1.18 curies of iodine-129 were contained in wastewater generated by the Idaho Chemical Processing Plant (ICPP) at the Idaho National Engineering Laboratory. The wastewater was discharged directly to the Snake River Plain aquifer through a deep disposal well prior to February 1984 and through unlined disposal ponds from 1984 to 1990. The wastewater did not contain measurable concentrations of iodine-129 in 1989–90.

In 1990–91, samples were collected from 51 wells that obtain water from the Snake River Plain aquifer and 1 well that obtains water from a perched ground-water zone. The samples were analyzed for iodine-129 using the accelerator mass spectrometer method, which is two to six orders of magnitude more sensitive than neutron-activation methods. Therefore, iodine-129 was detectable in samples from a larger number of wells distributed over a larger area than previously was possible. Ground-water flow velocities calculated using iodine-129 data are at least 6 feet per day. These velocities compare favorably with those of 4 to 10 feet per day calculated using tritium data and tracer studies at wells downgradient from the ICPP.

In 1990–91, concentrations of iodine-129 in water samples from wells that obtain water from the Snake River Plain aquifer ranged from less than 0.0000006 ± 0.0000002 to 3.82 ± 0.19 picocuries per liter (pCi/L). The mean concentration in water from 18 wells was 0.81 ± 0.19 pCi/L as compared with 1.30 ± 0.26 pCi/L in 1986. The decrease in the iodine-129 concentrations from 1986 to 1990–91 chiefly was the result of a decrease in the amount of iodine-129 disposed of annually and changes in disposal techniques.

INTRODUCTION

The U.S. Department of Energy's Idaho National Engineering Laboratory (INEL) includes about 890 mi² in southeastern Idaho (fig. 1). The INEL was established in 1949 to build, test, and operate different types of nuclear reactors. It also is a leading center for nuclear-safety research, defense programs, nuclear-waste technology, and advanced energy concepts. A key facility at the INEL is the Idaho Chemical Processing Plant (ICPP), which recovered uranium in spent fuel elements from government-owned reactors prior to 1993. As part of the fuel reprocessing activities, several fission products were released and were contained in wastewater at the ICPP. Iodine-129, produced by the fission of uranium-235 and plutonium-239, is one of the products released and contained in wastewater at the ICPP. Prior to

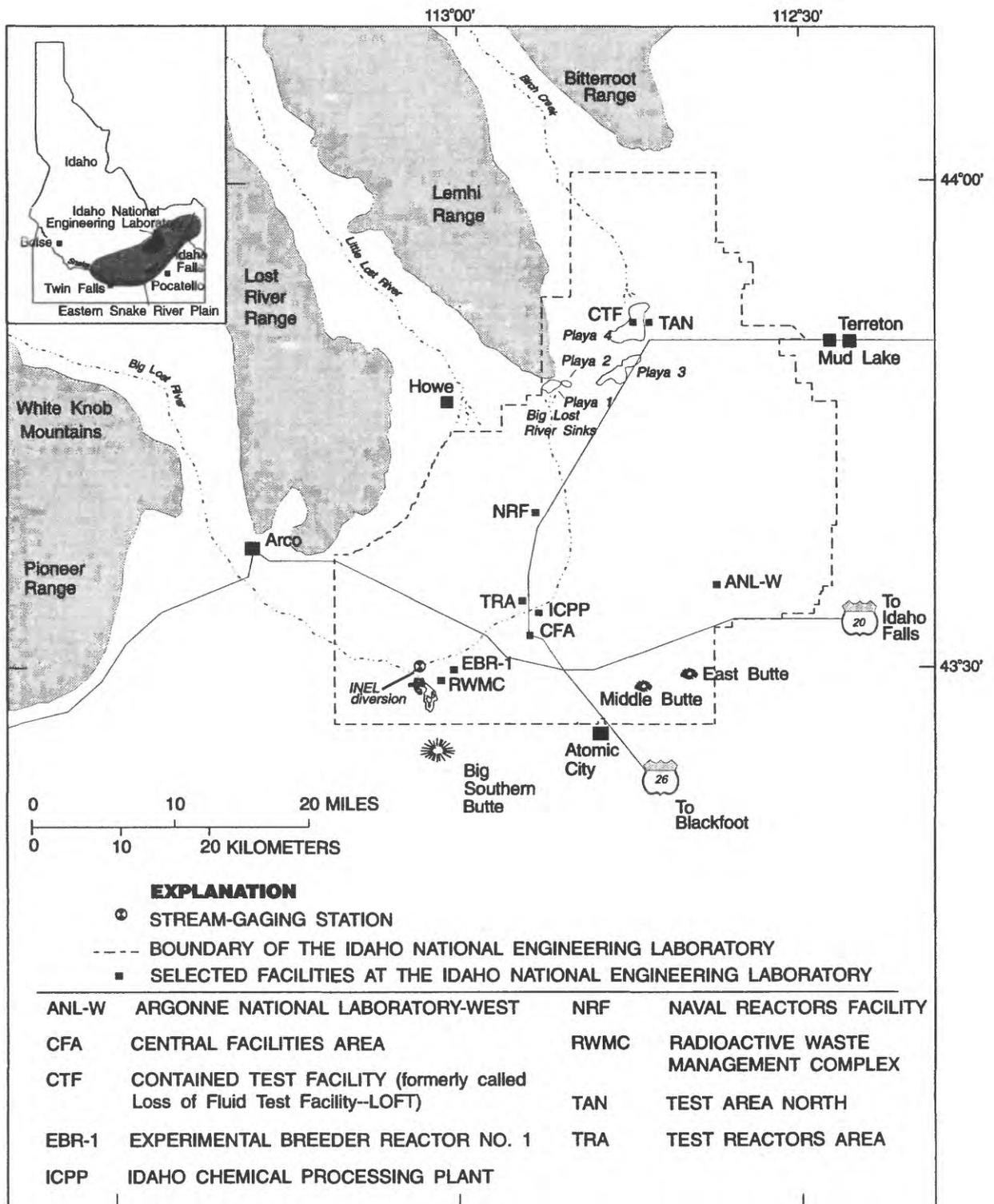


Figure 1.--Location of the Idaho National Engineering Laboratory.

1984, most of the wastewater generated at the ICPP was injected directly to the Snake River Plain aquifer through a 598-ft-deep disposal well. Beginning in February 1984, routine use of the disposal well was discontinued and wastewater was discharged to disposal ponds. The disposal ponds are unlined and wastewater infiltrates the pond bottoms and percolates through about 450 ft of underlying rock to the aquifer.

Because of its 15.7-million year radioactive half-life, iodine-129 is a permanent environmental pollutant. Since 1977, when wastewater monitoring for iodine-129 began, four sampling and analysis programs have been conducted to define the concentration and distribution of the radionuclide in water from the Snake River Plain aquifer. Results of the 1977, 1981, and 1986 programs were described by Mann and others (1988). This report describes the 1990–91 sampling and analysis program and changes in the concentration and distribution of iodine-129 in the aquifer since 1986. The program was conducted by the U.S. Geological Survey in cooperation with the U.S. Department of Energy's Environmental Measurements Laboratory (EML) in New York, New York and Idaho Operations Office in Idaho Falls, Idaho. Water samples were collected from 51 wells that obtain water from the aquifer and 1 well that obtains water from a perched ground-water zone at the ICPP.

The two major differences between the 1990–91 sampling and analysis program and previous programs are: (1) a larger number of wells were sampled; and (2) the ground-water samples were analyzed by the accelerator mass spectrometry (AMS) method instead of the neutron-activation method used for previous analyses. The method detection limit for AMS analyses is two to six orders of magnitude more sensitive than that for neutron-activation analyses.

Physical Setting

The INEL is on the eastern part of the Snake River Plain (fig. 1). Basaltic-lava flows intercalated with sedimentary deposits underlie the plain and make up the Snake River Plain aquifer, a major aquifer in Idaho. The aquifer provides the entire

water supply for the INEL and is a major source of irrigation, municipal, and industrial supplies for other parts of the plain. The depth to water in the aquifer ranges from about 200 ft below land surface in the northern part of the INEL to more than 900 ft in the southern part. At the ICPP, the depth to water is about 450 ft below land surface.

The general direction of ground-water movement at the INEL is to the southwest. Ground-water underflow in the aquifer at the INEL is maintained primarily by recharge in the northeastern part of the plain and from tributary drainages west and north of the INEL. From the recharge areas, the water moves beneath the INEL and eventually is discharged to springs along the Snake River near Twin Falls (Lewis and Jensen, 1985). Robertson and others (1974, p. 13) estimated the velocity of ground-water movement to be from 5 to 20 ft/day in the southern part of the INEL.

Streams at the INEL flow only in response to rainfall and snowmelt. The Big Lost River is the main drainage at the INEL and is in a topographically closed basin. Flow in the river is regulated by Mackay Dam—about 40 mi upstream from Arco—and by the INEL diversion which is in the southwestern part of the INEL (fig. 1). That part of the periodic streamflow that is not diverted flows northeast toward the ICPP and then north into the Big Lost River Sinks and a series of playas (fig. 1). A large part of the water that flows onto the INEL infiltrates permeable sedimentary deposits and basaltic rocks that underlie the river, sinks, and playas and recharges the Snake River Plain aquifer.

Previous Investigations

Many investigators have described the geology and hydrology of the Snake River Plain aquifer at the INEL. Robertson and others (1974) described the regional geology and hydrology and the influence of liquid waste disposal on the geochemistry of water for 1952–70. Barraclough and Jensen (1976) reported on hydrologic conditions from 1971–73. Barraclough and others (1982) provided an update on hydrological conditions for 1974–78; Lewis and Jensen (1985) provided an update for

1979–81; Pittman and others (1988), for 1982–85; and Orr and Cecil (1991), for 1986–88.

Distributions of iodine-129 in ground water were described by Barraclough and others (1982), Lewis and Jensen (1985), and Mann and others (1988).

Field Methods

Water samples from the Snake River Plain aquifer were collected at selected wells near and hydraulically downgradient from the ICPP and analyzed for iodine-129. Dedicated submersible or turbine pumps were used to obtain samples from the wells. Samples were collected in 1-L polyethylene bottles from a spigot in the discharge pipe. A thief sampler was used to collect a 1-L sample from the well that obtains water from a perched ground-water zone.

Water from wells equipped with dedicated pumps was monitored during sampling for temperature, pH, and specific conductance using methods described by Wood (1981) and Claassen (1982). A water sample was collected when these properties of the water stabilized and when a volume of water equivalent to a minimum of three well-bore volumes was pumped from each well; a well-bore volume is that volume of water in the interval of the well open to the aquifer plus the volume of water in ungrouted parts of the well bore and in the casing above the open interval. At many wells, 5 to 10 well-bore volumes were pumped prior to the collection of a sample. The diameter of the well bore, rather than the diameter of the casing, was used to calculate the minimum volume because of the potentially large difference between the two.

For the well that obtains water from a perched ground-water zone, a thief sampler was lowered inside the well casing to a predetermined level. The thief sampler is constructed so that water passes through the sampler while it is being lowered. At the predetermined level, the ends of the sampler are closed remotely, and about 1 L of water is trapped. Because the well was not pumped, the sample was collected opposite a fractured zone in the basaltic rocks, through which water likely moves at a high velocity compared with the velocity in unfractured zones; collecting a

sample opposite a fractured zone minimizes the likelihood of stagnated water in the well bore being sampled. The thief sampler was cleaned and rinsed with a pressurized spray of deionized water before and after use at the well. Methods used to collect iodine-129 samples prior to 1990 are summarized by Mann and others (1988).

Analytical Methods and Quality Assurance

Iodine-129 concentrations in the ground-water samples were determined using AMS methods described by Elmore and Phillips (1987). Accelerator mass spectrometers used to analyze the samples are at the IsoTrace Laboratory, University of Toronto, Toronto, Ontario, Canada, and at the Nuclear Structure Research Laboratory (NSRL), University of Rochester, Rochester, New York. The analyses were reported by the laboratories as the ratio of iodine-129 to iodine-127.

To ensure that the iodine-129 to iodine-127 ratios in ground-water samples did not exceed 10^{-10} , the upper limit of the AMS method, a combination of dilution and carrier-addition techniques was used during preparation of samples for analyses. Accordingly, 1:1, 1:10, 1:100, or 1:1,000 dilutions of sample water were prepared offsite using 18 megohm-centimeter water supplied by the EML. The dilutions and 1 L of sample water then were sent to EML where they were stored for approximately 2 weeks prior to continuing sample preparation. During the 2-week period, a low-background iodide carrier was prepared from a 20-percent weight-to-volume solution of potassium iodide (Lot AO-03) obtained from Anderson Laboratories, Inc.—tradename BANCO—of Fort Worth, Texas. This iodide is used at several AMS facilities and has been shown to deliver iodine-129 to iodine-127 ratios near 10^{-14} when the silver-iodide targets are prepared by direct precipitation using high-purity silver-nitrate reagent (Udo Fehn, University of Rochester, written commun., 1989, and L.R. Kilius, University of Toronto, written commun., 1990).

The iodide carrier was standardized as follows:
(1) An appropriate amount of the BANCO stock solution was diluted with 18 megohm-centimeter water to a concentration near 20 mg/mL using a

calibrated Eppendorf pipette and (2) six 1-mL aliquots of the carrier solution were titrated with a standardized silver-nitrate solution using eosin as an indicator (Furman, 1962). Iodide analyses of the solution in the six aliquots showed that the mean concentration and the standard deviation about the mean was 21.64 ± 0.04 mg/mL or 1.03×10^{20} atoms/mL.

After the carrier solution was prepared, all water samples were processed in a Class 100 clean room to ensure the radiochemical integrity of the samples. Initially, water samples from 12 of the 52 wells were selected and 10-mL aliquots of the 1:100 dilutions of each sample were chemically processed following the addition of 1 mL of the iodide carrier to each 10-ml aliquot. The quantity of iodide in each aliquot following the addition of the carrier greatly exceeded the concentration of stable iodide in the water from the Snake River Plain aquifer and provided ample silver iodide for the AMS targets; the iodide concentration in the aquifer generally is less than 0.001 mg/L. The iodine-129 and iodide carrier then were adjusted to the same valence state by adding 5 mL of reagent-grade sodium hypochlorite, which oxidizes I^- to IO_3^-/IO_4^- , and by allowing the resulting solution to stand overnight in a capped, 50-mL polyethylene centrifuge tube. The solution then was heated to approximately 70°C for 1 hour in a water bath to further ensure oxidation/equilibration before proceeding with sample preparation. Thereafter, the procedure of Glendenin and Metcalf, as detailed by Kleinberg and Cowan (1960), was used to isolate iodine. All chemical manipulations were performed in the single, original 50 mL polyethylene centrifuge tube to avoid the possibility of cross-contamination between samples. The purified iodine was precipitated from a dilute acid solution as silver iodide using an ultra-pure silver nitrate solution. The precipitate was collected by centrifugation, transferred to pre-cleaned 1 dram amber glass vials with inert caps, dried overnight at 100°C, and mailed to the laboratories for analysis.

The AMS measurements of iodine-129 in the first 12 samples indicated that 1-mL aliquots of sample water would yield acceptable iodine-129 to iodine-127 ratios and, thereafter, the majority of the samples were processed using this sample

volume Reagent blanks consisted of 1 to 10 mL of the 18 megohm-centimeter water that also was used for sample dilutions plus 1 mL of iodide carrier.

At IsoTrace, all targets were prepared by adding a niobium metal binder (an ultra-pure silver metal binder was used at the NSRL) to the purified silver iodide in a ratio of 1:1 and baking the resultant mixture for 6 hours or more at 100°C in a vacuum oven; stainless-steel sample holders were used to position the targets in the ion source of the accelerator. Negative ion currents greater than 3 microamperes are routine from such sample preparations (Kieser, 1989). A more detailed description of the AMS iodine-129 program at IsoTrace is described by Kilius and others (1987); Kubik and others (1987) describe the program at the NSRL.

Iodine-129 to iodine-127 ratios and associated analytical uncertainties were provided by the laboratories for each target submitted for analysis. From these ratios and uncertainties, the concentrations and analytical uncertainties of iodine-129 in picocuries per liter were calculated by the authors. For each iodine-129 concentration, an associated analytical uncertainty, s , was calculated so that there was a 67-percent probability that the true concentration in a sample was in the range of the reported concentration plus or minus the analytical uncertainty. For example, given an analytical result of 1.68 ± 0.05 pCi/L, there is a 67-percent probability that the true concentration is in the range of 1.63 to 1.73 pCi/L; at 2s or 1.68 ± 0.10 pCi/L, there is a 95-percent probability that the true concentration is in the range of 1.58 to 1.78 pCi/L.

Quality assurance and reproducibility of the iodine-129 measurements were tested in three ways: (1) An iodine-129 standard with known activity was used to calibrate the AMS unit for iodine-129 measurements; (2) seven water samples were analyzed as blind replicates at IsoTrace; and (3) four replicate water samples were analyzed at the NSRL. Both the IsoTrace Laboratory and NSRL calibrate their iodine-129 measurements using dilutions prepared from the National Institute of Standards and Technology standard reference material #4949b, a calibrated iodine-129 solution

of known activity. At IsoTrace, serial, volumetric dilution of standard reference material and appropriate addition of stable iodide carrier were used to prepare an internal laboratory standard with a theoretical iodine-129 to iodine-127 ratio of $(1.1739 \pm 0.0331) \times 10^{-11}$. The theoretical ratio of the internal laboratory standard compared favorably to its measured ratio of $(1.1789 \pm 0.0211) \times 10^{-11}$ when run against an iodine-129 to iodine-127 standard supplied by Dr. June Fabryka-Martin of Los Alamos National Laboratory (Kilius, 1990). When measured against the NSRL iodine-129 to iodine-127 standard, the IsoTrace standard produced a ratio of $(1.3127 \pm 0.1313) \times 10^{-11}$, which is in reasonable agreement with the IsoTrace determination given that the uncertainties in the measurements overlap at the 67-percent confidence interval.

Analyses of blind replicate samples generally were in agreement (see table 2 in section entitled "Concentrations of Iodine-129 in the Snake River Plain aquifer"). Most of the reported results for those samples which were run at both IsoTrace and the NSRL overlap at the 67-percent confidence interval when the uncertainties in the measurements are taken into account. Exceptions are results for samples from wells 36 and 67 (replicates analyzed at IsoTrace); and samples from CFA-2 for which the IsoTrace and NSRL results differ by nearly a factor of 2.

Differences in the AMS measurements of iodine-129 are attributed to three factors: (1) improper equilibration of the iodide carrier added to the sample prior to chemical processing; (2) cross-contamination of samples during processing or in the ion source of the accelerator during the sputtering process; and (3) variations that occur during the sputtering process between samples and the standards used to quantitate the iodine-129 to iodine-127 ratios in the respective samples. Of these possibilities, factors 1 and 2 are most likely to lead to differences of the magnitude reported for well CFA-2; smaller differences between replicates, comparable to those for wells 36 and 67, might reasonably be attributed to factor 3. Because sample targets are loaded onto a sample wheel that also contains standards (blanks are analyzed on separate wheels), there is a

potential that high-level samples with iodine-129 to iodine-127 ratios greater than or equal to 10^{-9} are loaded onto a wheel where the standard is 10^{-11} . In such instances, the possibility exists that small amounts of sample can contaminate the surface of the standard and vice versa during the sputtering process. Because the magnitude of errors caused by cross contamination is difficult to quantify, samples with comparable iodine-129 to iodine-127 ratios are analyzed as a group whenever possible. Moreover, variations that occur during the sputtering and acceleration process between samples and standards cannot be accounted for in AMS measurements of iodine-129. Unlike AMS measurements of chlorine-36 and carbon-14, iodine-127 is the only stable isotope of iodine; "fractionation" effects that occur during the analysis between standards and samples cannot be normalized by reference to ratio differences measured for the stable isotope counterparts as can be done for chlorine-36 and carbon-14 (Suter, 1990). It should be emphasized, however, that differences in the iodine-129 to iodine-127 ratios for replicate analyses greater than 10 percent, most likely result from factors 1 and 2.

Guidelines for the Interpretation of Analytical Results

Concentrations of iodine-129 are reported with an estimated sample standard deviation, s , that is obtained by propagating sources of analytical uncertainties in measurements. The following guidelines for interpreting analytical results are based on an extension of the method described by Currie (1968).

In the analysis for iodine-129, laboratory measurements are made on a target sample and a prepared blank. Instrument signals for the sample and the blank vary randomly. Therefore, it is essential to distinguish between two key aspects of the problem of detection: (1) The instrument signal for the sample must be greater than the signal for the blank to make the decision that iodine-129 was detected; and (2) an estimation must be made of the minimum concentration that will yield a sufficiently large observed signal to make the correct decision for detection or

nondetection of iodine-129. The first aspect of the problem is a qualitative decision based on an observed signal and a definite criterion for detection. The second aspect of the problem is an estimation of the detection capabilities of a given measurement process.

In the laboratory, instrument signals must exceed a critical level to make the decision whether iodine-129 was detected. Iodine-129 concentrations that equal 1.6s meet this criterion; at 1.6s, there is a 95-percent probability that the correct conclusion—not detected—will be made. Given a large number of samples, as many as 5 percent of the samples with measured concentrations greater than or equal to 1.6s, which were concluded as being detected, might not contain iodine-129. These measurements are referred to as false positives and are errors of the first kind in hypothesis testing.

Once the critical level of 1.6s has been defined, the minimum detectable concentration can be established. Iodine-129 concentrations that equal 3s represent a measurement at the minimum detectable concentration. For true concentrations of 3s or greater, there is a 95-percent or more probability of correctly concluding that iodine-129 was detected in a sample. Given a large number of samples, the conclusion—not detected—will be made for up to 5 percent of the samples with true concentrations greater than or equal to 3s. These measurements are referred to as false negatives and are errors of the second kind in hypothesis testing.

True iodine-129 concentrations between 1.6s and 3s have larger errors of the second kind. That is, there is a greater than 5-percent probability of false negative results for samples with true concentrations between 1.6s and 3s; at 1.6s, the probability of a false negative is about 50 percent.

These guidelines are based on counting statistics alone and do not include systematic or random errors inherent in laboratory procedures. The values, 1.6s and 3s, vary slightly with background or blank counts, and with the number of gross counts for individual analyses. The use of the critical level and minimum detectable concentration aid in the interpretation of analytical

results and do not represent absolute concentrations of radioactivity which may or may not have been detected. In this report, if the iodine-129 concentration was less than 3s, the concentration is considered to be below a "reporting level." The reporting level should not be confused with the analytical method detection limit, which is based on laboratory procedures.

Occasionally, an iodine-129 concentration will be negative—for example -0.003 ± 0.004 pCi/L. One or two "blank" samples are prepared and analyzed with each group of water samples analyzed by AMS method to define background conditions that are inherent in target preparation as well as those in the laboratory. As part of the calculations involved in converting the iodine-129 to iodine-127 ratios to iodine concentrations in picocuries per liter, the ratio for the blank samples is subtracted from the ratio for a water sample. If the ratio for the blank sample is greater than the ratio for the ground-water sample, the resultant difference will be negative.

Acknowledgments

The authors gratefully acknowledge Dr. Linas R. Kilius of the IsoTrace Laboratory and Drs. Pankaj Sharma and Peter Kubik of the NSRL, for their interest in this research and for their advice, counsel, and personal efforts to ensure the quality of data in this report. Additionally, we wish to thank U.S. Geological Survey hydrologists R.J. Edmonds, Tempe, Ariz., and W.W. Staubitz, Tacoma, Wash. for their constructive comments, which improved the clarity and technical content of this report.

OCCURRENCE AND CHARACTERISTICS OF IODINE-129

Iodine-129 is a naturally occurring radioisotope primarily produced by the spontaneous fission of uranium-238, reaction of cosmic radiation with xenon isotopes and cosmic dust in the upper atmosphere, neutron capture reactions with tellurium, and fission of actinides. Fission contributions of iodine-129 to the hydrosphere originate from uranium-238 in seawater and weathering rocks,

and through the release of iodine-129 in volcanic processes. Naturally occurring iodine-129 in the environment was estimated to be about 40 Ci by the National Council on Radiation Protection and Measurements (1983). However, data provided by Fabryka-Martin and others (1985) indicate that iodine-129 in the geosphere is more than two orders of magnitude greater than that estimated by the National Council on Radiation Protection and Measurements. Global inventories predicted for iodine-129 are about 54.3 million grams or 9,600 Ci, of which about 52 million grams or 9,200 Ci are associated with igneous activity (Fabryka-Martin and others, 1985, fig. 1).

Iodine-129 also is produced by the fission of uranium-235 and plutonium-239. Measurable amounts were produced by the atmospheric testing of nuclear weapons and in the production of electricity by nuclear-power plants. The largest source of iodine-129 is spent nuclear fuels; however, iodine-129 can be released only when the irradiated fuel is reprocessed. The approximate releases of iodine-129 from atmospheric and high-altitude nuclear weapons tests to 1975 totaled about 10 Ci; the estimated inventory in spent fuel from commercial reactors in the United States through 1982 was about 180 Ci (National Council on Radiation Protection and Measurements, 1983), although only a small part of this iodine-129 has been released during reprocessing.

The ICPP reprocessed spent nuclear fuel used in government reactors prior to 1993. Between 1953, when the ICPP began operations, and 1990, it is estimated that 0.56 to 1.18 Ci of iodine-129 were contained in wastewater discharged to the disposal well and ponds (White, 1977–78; O.L. Cordes, Allied Chemical Corp., written commun., 1978; and EG&G Idaho, Inc., 1979–91). Iodine-129 in wastewater at the ICPP accounts for nearly all of the radionuclide in wastewater at the INEL (Mann and others, 1988, p. 9).

During 1987–90, iodine-129 concentrations in wastewater discharged to the disposal ponds were less than those in wastewater previously discharged to the disposal well and ponds. The reduction is the result of: (1) the waste stream containing iodine-129 is now recycled and

ultimately will be stored with high-level radioactive waste; and (2) in 1989–90, no fuel was processed at the ICPP (G.A. Hula, U.S. Department of Energy, written commun., 1992).

Concentrations of Iodine-129 in the Snake River Plain Aquifer

In 1990–91, water samples were collected from 51 wells that obtain water from the Snake River Plain aquifer at and adjacent to the INEL. Additionally, a sample was collected from one well at the ICPP that obtains water from a perched ground-water zone to determine whether water in the zone could be a source of iodine-129 to the aquifer. The depths of wells, intervals of wells open to the aquifer, depths to water, and dates water samples were collected are shown on table 1. Concentrations of iodine-129 in water from the Snake River Plain aquifer ranged from 0.0000006 ± 0.0000002 to 3.82 ± 0.19 pCi/L and some were less than the reporting level. The reporting level differed from one sample to another depending on the amount of dilution and size of aliquot used to prepare the target for analysis by AMS. For example, the target prepared using water from well 65 was prepared with a 10-mL aliquot of sample water that was diluted 1:10, whereas the target for well 83 was prepared using 1 L of undiluted sample water; the concentration in water from well 65 was 0.0008 ± 0.0040 pCi/L—less than the reporting level—but the concentration in water from well 83 was 0.0000006 ± 0.0000002 pCi/L and is considered a detectable concentration. Concentrations of iodine-129 in water samples from selected wells in 1990–91 are shown on table 2.

In general, iodine-129 concentrations decreased between 1986 and 1990–91. The decrease was a continuance of the 1981 to 1986 decrease described by Mann and others (1988, p. 16). The 1981–86 decrease in iodine-129 concentrations largely is attributed to changes in disposal techniques, but also resulted from decreases in the annual amount of iodine-129 contained in wastewater, sorption, and dilution by recharge from the infiltration of streamflow in the Big Lost River. The general decrease in iodine-129 concentrations

Table 1.--Depths of wells, intervals of wells open to aquifer, depths to water and dates water samples were collected for iodine-129 analyses, Idaho National Engineering Laboratory

[See Figures 5, 6, and 7 for well locations. All wells except well 50 obtain water from the Snake River Plain aquifer; well 50 obtains water from a perched ground-water zone at the Idaho Chemical Processing Plant]

Well identifier	Depth of well (feet)	Interval of well open to aquifer (feet)		Depth to water (feet)	Date measured	Date sampled
11	704	673-704		653	8/21/91	8/21/91
14	751	715-751		716	8/21/91	8/21/91
20	676	471-481	512-553	460	9/10/90	9/10/90
34	700	499-700		471	8/23/90	8/23/90
35	578	145-578		472	8/23/90	8/23/90
36	567	430-567		471	8/23/90	8/23/90
37	572	507-572		471	7/5/90	7/5/90
38	729	678-729		472	10/5/90	10/5/90
39	572	47-572		473	8/23/90	8/23/90
40	483	456-483		458	7/5/90	7/5/90
41	674	428-674		458	8/30/90	8/30/90
42	678	453-678		457	8/30/90	8/30/90
43	676	451-676		457	7/25/90	7/25/90
44	650	461-650		459	7/26/90	7/26/90
45	651	461-651		460	7/26/90	7/27/90
46	651	461-651		458	7/26/90	7/27/90
47	651	460-651		455	7/24/90	7/24/90
48	750	462-750		458	9/13/90	9/13/90
50	405	357-405		376	7/24/90	7/24/90
51	659	475-659		461	10/10/90	7/26/90
52	650	450-650		450	8/30/90	8/30/90
57	732	477-732		464	6/28/90	6/28/90
59	657	464-657		454	7/24/90	7/24/90
65	498	456-498		464	7/11/90	7/11/90
67	698	465-552	635-698	456	7/25/90	7/25/90
77	610	470-610		464	7/26/90	7/26/90
82	700	470-570	593-700	448	7/26/90	3/27/91
83	752	516-752		498	8/15/91	8/15/91
85	637	522-637		482	9/10/90	9/10/90
88	662	587-662		587	7/19/90	7/19/90

Table 1.--Depths of wells, intervals of wells open to aquifer, depths to water and dates water samples were collected for iodine-129 analyses, Idaho National Engineering Laboratory--Continued

Well identifier	Depth of well (feet)	Interval of well open to aquifer (feet)		Depth to water (feet)	Date measured	Date sampled
89	646	576-646		601	7/2/90	7/2/90
90	626	580-626		582	7/2/90	7/2/90
103	760	575-760		583	7/9/90	7/9/90
104	700	550-700		555	7/9/90	3/28/91
105	800	400-800		670	9/28/90	3/28/91
106	760	400-760		587	6/27/90	6/27/90
108	760	400-760		608	10/24/90	3/28/91
111	600	440-600		468	6/26/90	6/26/90
112	563	432-563		472	7/11/90	7/11/90
113	564	445-564		473	6/26/90	6/26/90
114	562	440-562		467	6/27/90	6/27/90
115	581	440-581		463	7/9/90	7/9/90
116	580	400-580		457	6/27/90	6/27/90
121	475	449-475		452	3/25/91	3/25/91
122	480	449-475		456	3/25/91	3/25/91
123	481	450-475		461	3/25/91	3/25/91
CFA-1	639	444-639		Not measured		7/11/90
CFA-2	681	521-651	661-681	474	1/25/89	7/11/90
ICPP-1	585	460-485	527-577	Not measured		7/24/90
ICPP-2	605	458-483	551-600	Not measured		7/24/90
ICPP Disposal	598	412-452	490-593	Well destroyed	10/16/89	
ICPP-4	700	450-700		441	10/17/83	7/23/90
TRA Disposal	1,270	512-697	930-1,070			
		1,183-1,268		464	7/16/90	7/16/90

Table 2.--Iodine-129 concentrations in ground water, Idaho National Engineering Laboratory, 1986 and 1990-91

[See figures 5 and 7 for well locations, and table 1 for selected information about the wells and the dates the 1990-91 samples were collected. Concentrations and analytical uncertainties are in picocuries per liter; uncertainties are 1s (67-percent confidence level). Data for 1986 are from Mann and others (1988, table 1). The 1990-91 water samples were analyzed by the IsoTrace Laboratory, University of Toronto, Toronto, Ontario, except as noted in the remarks column. Abbreviations: RL, concentration is less than the 3s reporting level; T, sample collected with a thief sampler; P, sample collected from a piston pump installed in tandem with a submersible pump. Remarks (applicable only to the 1990-91 analyses): 1991, sample analyzed in 1991; NSRL, sample analyzed by the Nuclear Structure Research Laboratory, University of Rochester, Rochester, New York]

Well identifier	1986	1990-91	Remarks
11	--	0.000010±0.000001	
14	--	.000030±0.000002	
20	0.1±0.3 (RL, T)	.033±0.002	
34	.73±0.15 (T)	.39±0.02	
35	1.0±0.2 (T)	.57±0.03	
36	2.0±0.2 (T)	1.19±0.03	
36	--	1.74±0.06	Blind replicate
36	--	1.17±0.05	
37	2.9±0.3 (T)	1.80±0.08	
38	.1±0.1 (RL, T)	2.00±0.07	
38	--	2.10±0.07	Blind replicate
39	.00±0.13 (RL, T)	.005±0.001	
40	1.4±0.2	.50±0.02	
41	.4±0.2 (RL, T)	.58±0.03	
42	.2±0.2(RL, T)	3.82±0.19	
43	.17±0.16 (RL)	.16±0.01	
44	.4±0.3 (RL, T)	.20±0.03	
45	1.1±0.2 (T)	.32±0.01	
45	--	.26±0.02	Blind replicate
45	--	.24±0.01	1991
45	--	.23±0.01	1991, blind replicate
46	2.3±0.3 (T)	.35±0.02	
47	1.5±0.2	.83±0.04	
48	.5±0.2 (RL, T)	.22±0.02	
50	--	1.20±0.04 (T)	Water from a perched ground-water zone

Table 2.--Iodine-129 concentrations in ground water, Idaho National Engineering Laboratory, 1986 and 1990-91--Continued

Well identifier	1986	1990-91	Remarks
51	.57±0.14 (T)	.28±0.01	
52	.9±0.2 (T)	.38±0.03	
57	3.6±0.4 (T)	1.36±0.07	
57	--	1.41±0.04	Blind replicate
59	.3±0.2 (RL, T)	.57±0.02	
65	--	.0008±0.0040 (RL)	
65	--	.004±0.005 (RL)	NSRL
67	2.4±0.3	1.43±0.04	
67	--	1.68±0.05	Blind replicate
67	--	1.59±0.08	1991
77	2.9±0.3 (T)	1.37±0.06	
82	1.6±0.3 (T)	.119±0.002	
83	--	.0000006±0.0000002	
85	3.3±0.3 (T)	1.64±0.08	
88	--	.00003±0.00004 (RL, P)	
88	--	.00004±0.00005 (RL, P)	NSRL
88	--	.00001±0.00004 (RL)	
89	--	-0.003±0.004 (RL)	
90	--	.0089±0.0004	
103	--	.00010±0.00004 (RL)	
103	--	.00008±0.00005 (RL)	NSRL
104	--	.0036±0.0001	1991
105	--	.00053±0.00004	1991
106	--	.025±0.001	
108	--	.00083±0.00002	1991
111	2.8±0.3	.86±0.09	
112	3.3±0.3	2.40±0.25	
113	2.9±0.3	3.25±0.14	
114	0.3±0.2 (RL)	.28±0.01	
115	.25±0.16 (RL)	.19±0.02	

Table 2.--Iodine-129 concentrations in ground water, Idaho National Engineering Laboratory, 1986 and 1990-91--Continued

Well identifier	1986	1990-91	Remarks
116	.65±0.16	.45±0.01	
121	--	.0000009±0.0000002	
122	--	.37±0.02	
123	--	1.00±0.05	
CFA-1	.49±0.12	.24±0.05	
CFA-1	--	.28±0.02	1991
CFA-2	-0.15±0.13 (RL)	.10±0.03	
CFA-2	--	.18±0.01	NSRL
ICPP-1	.5±0.3 (RL)	.033±0.010	
ICPP-2	.1±0.2 (RL)	.015±0.007 (RL)	
ICPP-2	--	.005±0.004 (RL)	1991
ICPP-4	.0±0.2 (RL)	-0.0001±0.0030 (RL)	
ICPP-4	--	.0005±0.0030 (RL)	Blind replicate
TRA Disposal	--	.011±0.001	1991

for 1981–86 was greater than the decrease from 1986 to 1990–91. The maximum concentration in 1990–91 was not markedly different from the maximum in 1986 and may have increased slightly. In 1986, the maximum concentration was 3.6 ± 0.4 pCi/L in water from well 57; in 1990, the maximum concentration was 3.82 ± 0.19 pCi/L in water from well 42 (table 2). The increase in the maximum concentration may be attributed to changes in the field sampling methods. In 1986, many of the water samples were collected using a thief sampler. In 1990–91, however, all wells that obtain water from the Snake River Plain aquifer were equipped with dedicated pumps and samples were collected from discharge pipes following extensive purging of the wells. Of the 20 wells in which dedicated pumps were installed between 1986 and 1990, the concentration of iodine-129 increased in samples from 4 of these wells and decreased in the others (table 2).

The general decrease in iodine-129 concentrations from 1986 to 1990–91 is attributed to some of the same factors responsible for the 1981-to-1986 decrease. The annual amount of iodine-129 contained in wastewater decreased markedly from 1986 to 1990–91. However, dilution by recharge from the infiltration of streamflow in the Big Lost River, did not have as great an effect; there was no flow in the river from 1987 to 1990–91. The iodine-129 concentrations in wastewater discharged to the disposal well and ponds at the ICPP from 1976 to 1990 are shown on figure 2 and annual discharge in the Big Lost River below INEL diversion from 1965 to 1990 is shown on figure 3.

Of the 51 wells sampled in 1990–91, 18 wells were sampled in 1977, 1981, and 1986. The mean concentration of iodine-129 in water from these 18 wells decreased between 1986 and 1990–91. The mean concentration and its experimental standard error was 1.30 ± 0.26 pCi/L in 1986 as compared with 0.81 ± 0.19 pCi/L in 1990–91. For purposes of comparison, the mean and experimental standard error of the mean for iodine-129 concentrations in water from the 18 wells are shown on figure 4; the 18 wells are 20, 37, 40, 42, 43, 45, 47, 51, 52, 57, 59, 67, 77, 82, 85, CFA-1 and -2, and ICPP-2.

Areal Distribution of Iodine-129 in the Snake River Plain Aquifer

In 1990–91, concentrations of iodine-129 in the Snake River Plain aquifer were greater than 0.25 pCi/L in a 3.4-mi² area (fig. 5). In contrast, water in the aquifer contained more than 0.25 pCi/L of iodine-129 in an area of about 4.3 mi² in 1986 (fig. 6). Similarly, the area in which the aquifer contained more than 1 pCi/L of iodine-129 in 1990–91 was about 1.5 mi² as compared with 2.5 mi² in 1986. In 1990–91, concentrations greater than 3 pCi/L were detected in water from well 42 which is about 0.25 mi southwest of the ICPP disposal well and well 113, which is about 1.2 mi south; in 1986, water from wells 57, 85, and 112 contained concentrations greater than 3 pCi/L (table 2).

Because the method detection limit for water samples collected in 1990–91 was from two to six orders of magnitude more sensitive than that for samples previously collected, iodine-129 was detected in water from a larger number of wells distributed over a larger area. Therefore, it was necessary to estimate the background concentration of iodine-129 produced naturally and by atmospheric testing of nuclear weapons to ascertain whether small concentrations at some wells resulted from activities at the ICPP.

Well 121 was drilled and constructed specifically to provide information on background concentrations for constituents which have contaminated or potentially could contaminate ground water at the ICPP. It is about 0.5 mi hydraulically upgradient and across the Big Lost River from the ICPP (fig. 7). It is unlikely that wastewater disposal at the ICPP has had a measurable effect on the concentration of iodine-129 in water from well 121 owing to recharge from the Big Lost River and the distance and direction of the well from ICPP wastewater-disposal facilities. The 0.0000009 ± 0.0000002 pCi/L of iodine-129 in water from well 121 likely is the result of natural production and the atmospheric testing of nuclear weapons; water from well 121 undoubtedly contains iodine-129 generated during the atmospheric testing program that was contained in recharge from the Big Lost

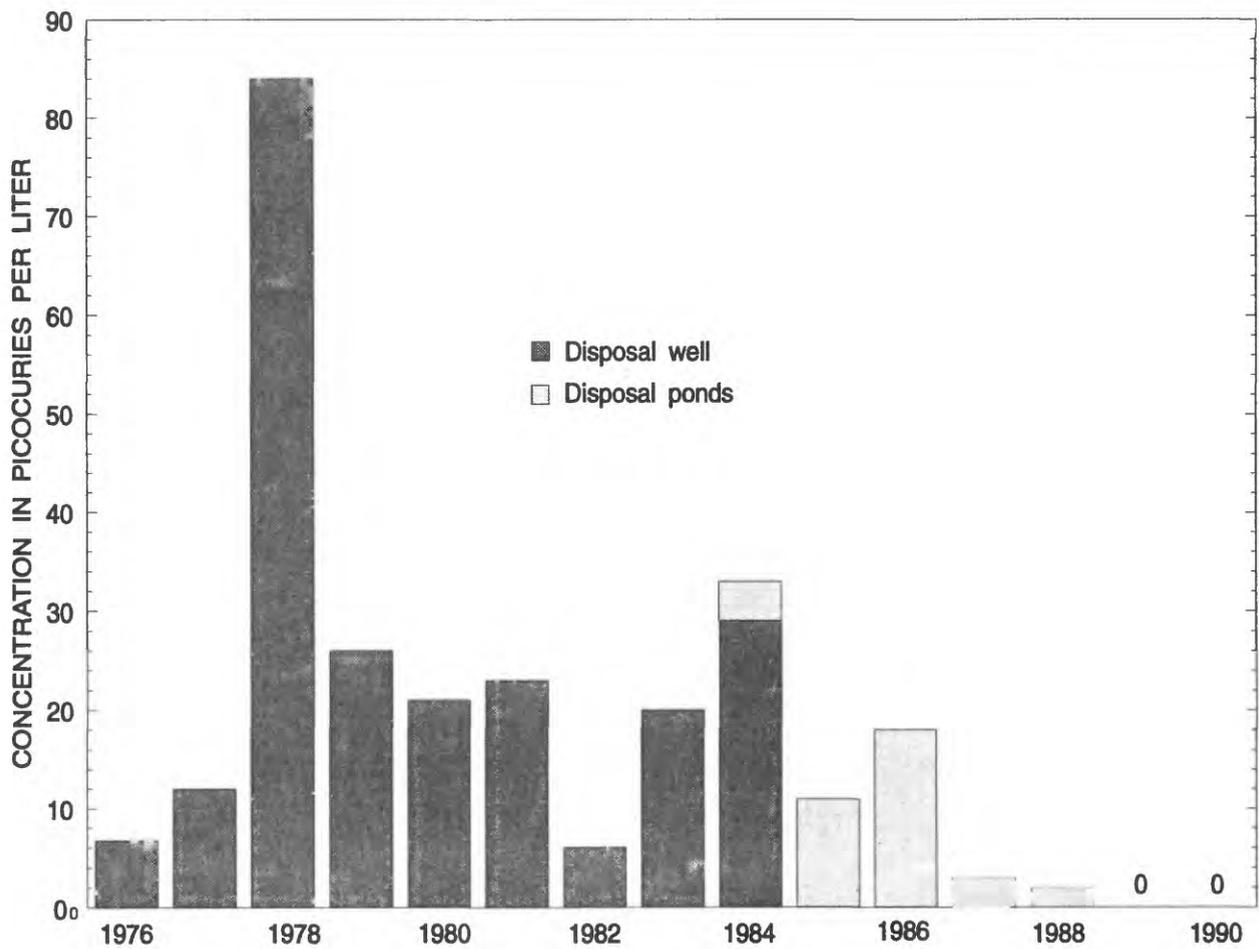


Figure 2.--Mean annual concentrations of iodine-129 in wastewater discharged to the disposal well and ponds at the Idaho Chemical Processing Plant [data from White (1977, 1978) and EG&G Idaho, Inc. (1979-91)].

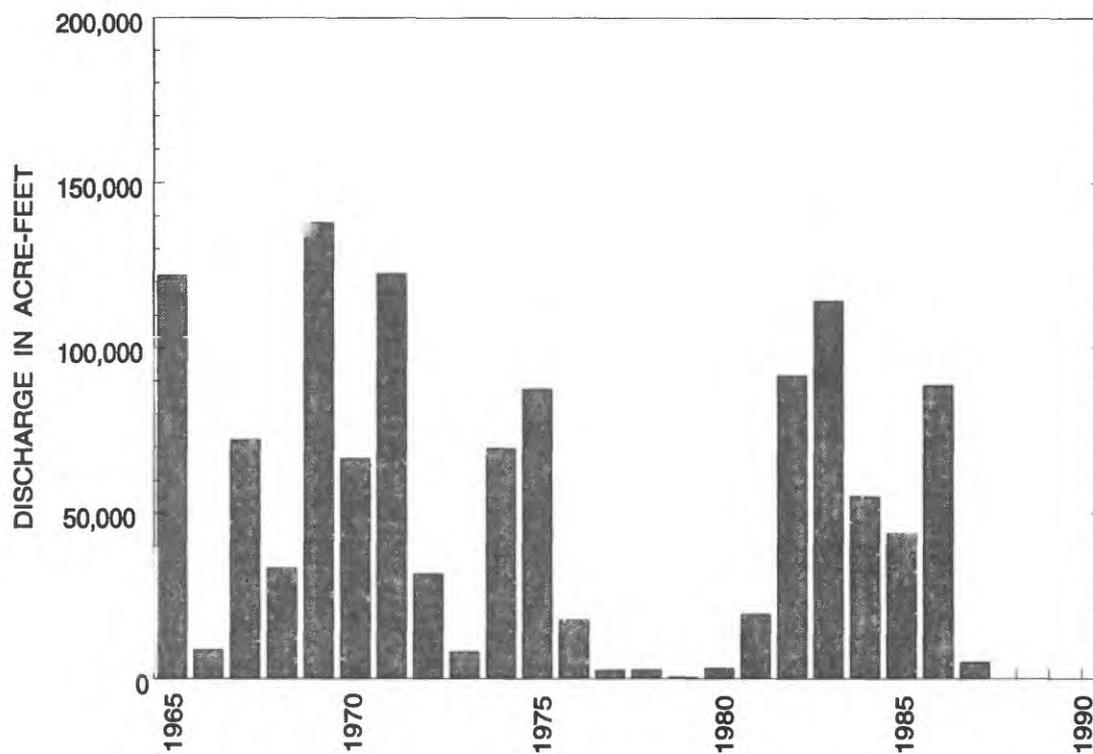


Figure 3.--Annual discharge of the Big Lost River below INEL diversion [data from Harenberg and others (1989-91) and Bennett (1990)].

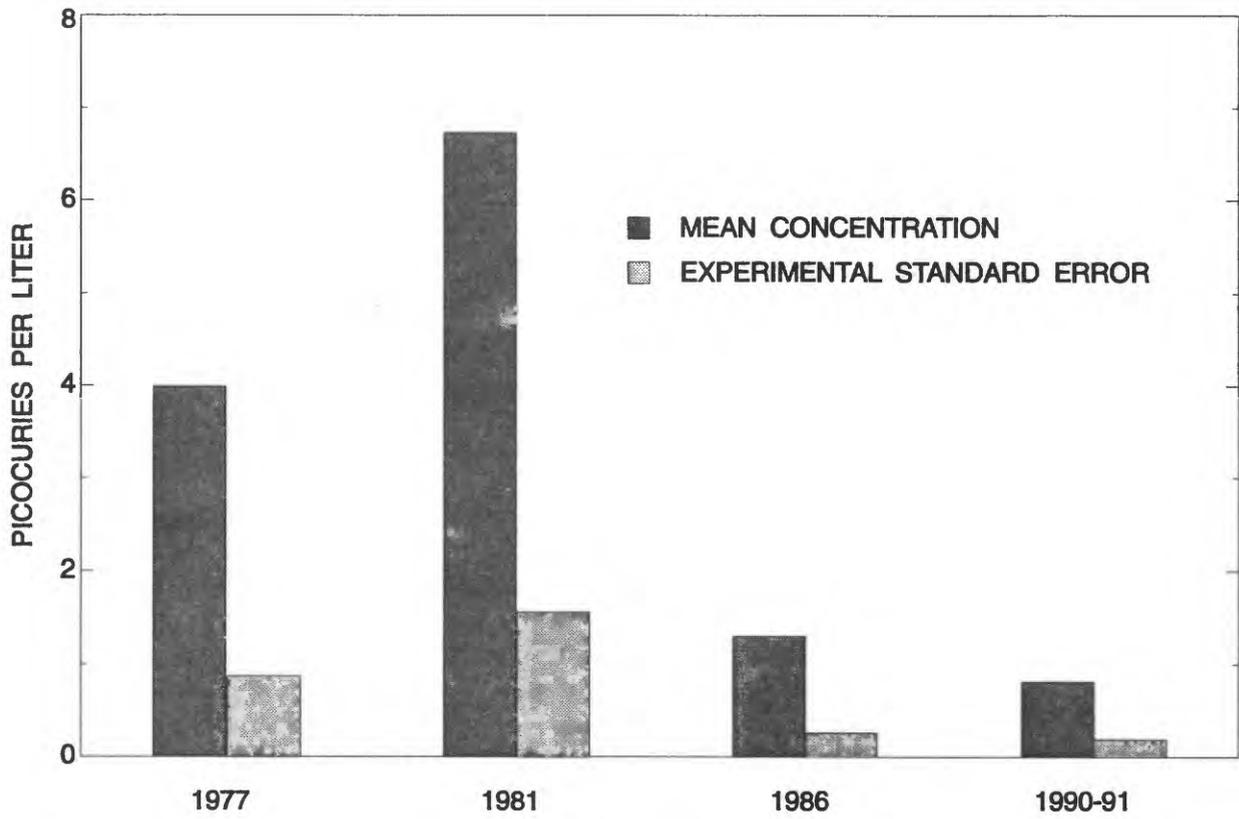
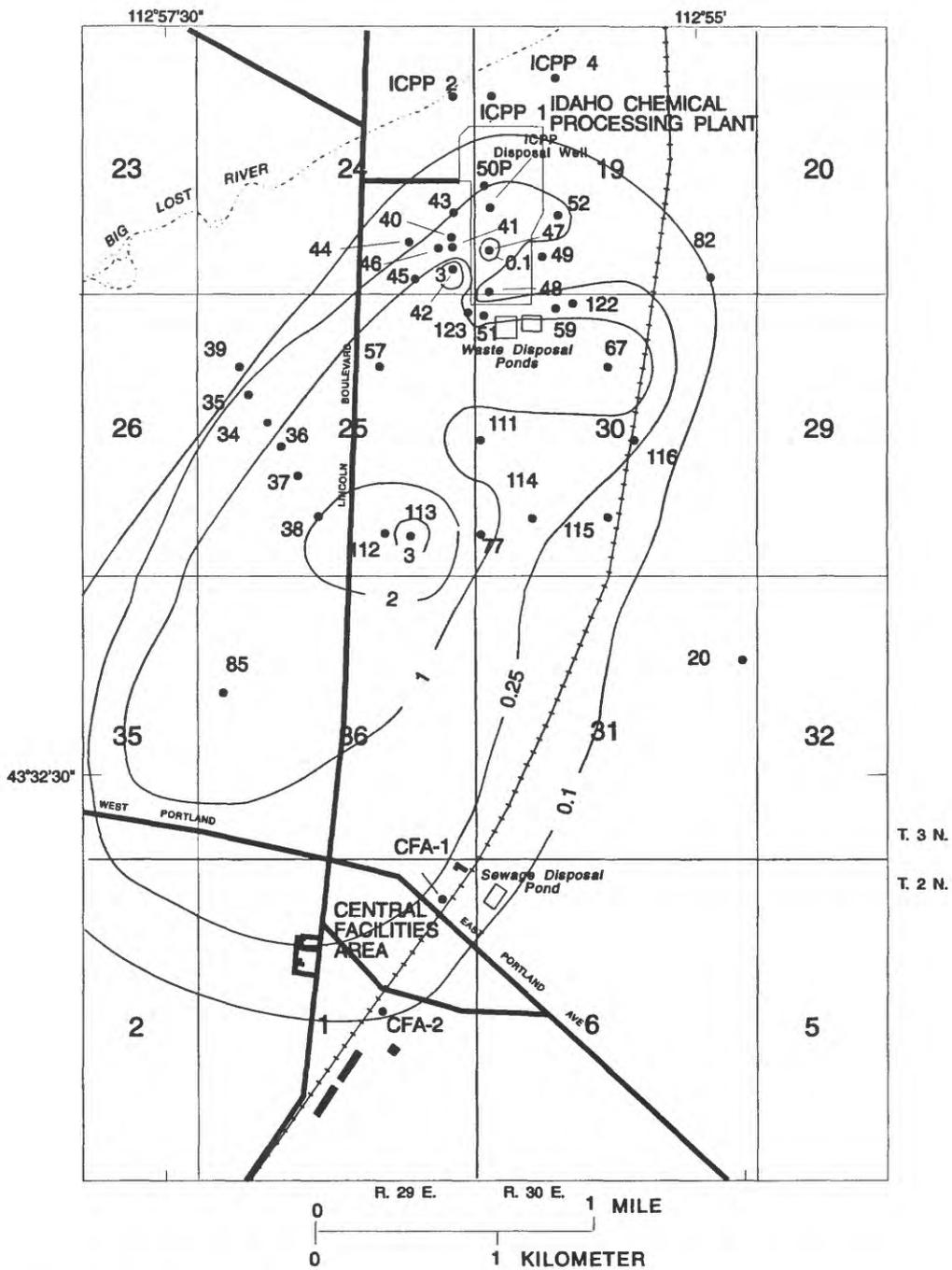


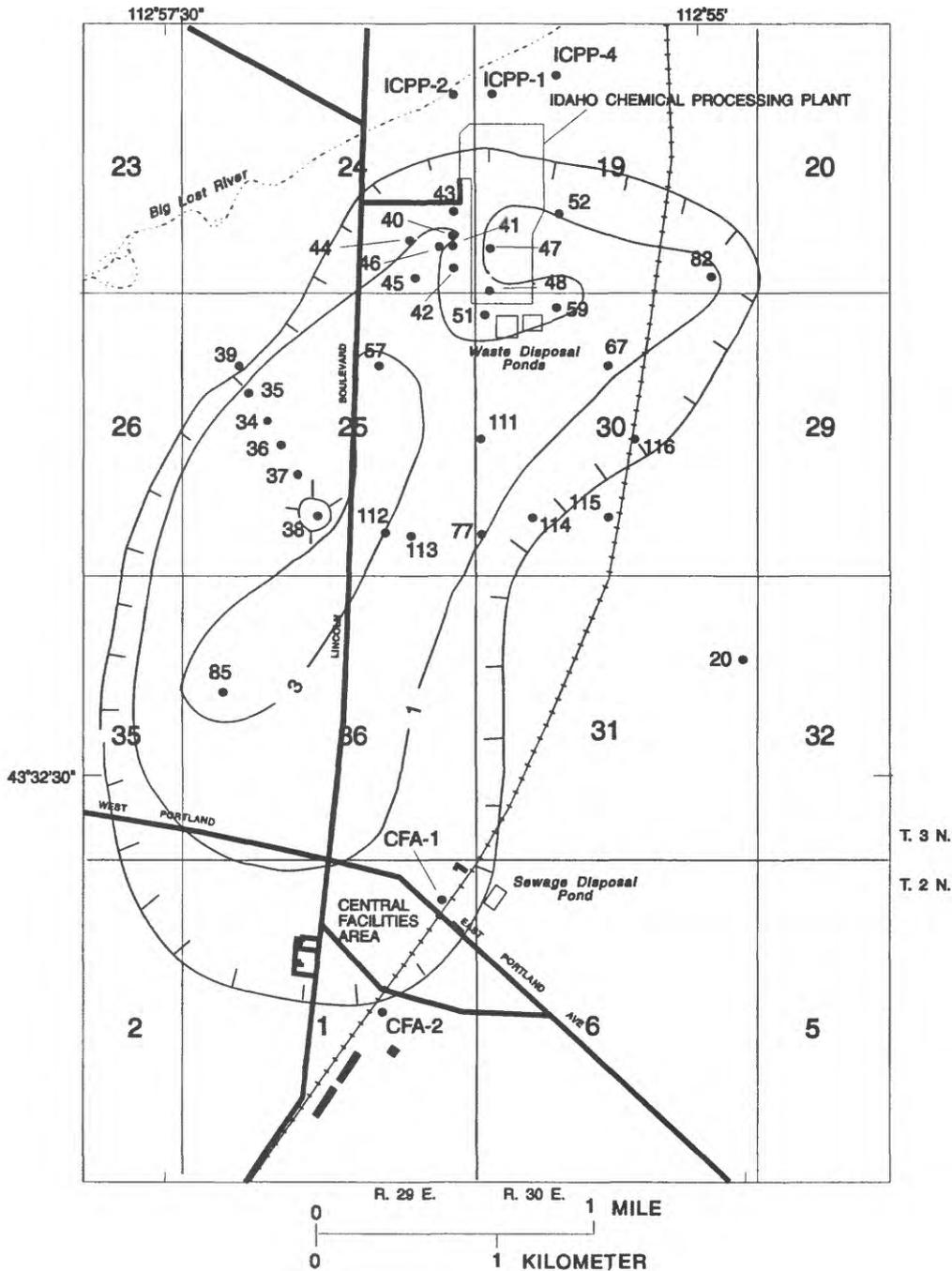
Figure 4.--Mean concentrations of iodine-129 and the experimental standard errors of the means in water from 18 wells sampled in 1977, 1981, 1986, and 1990-91, Idaho National Engineering Laboratory.



EXPLANATION

- 1 — LINE OF EQUAL IODINE-129 CONCENTRATION--Interval, in picocuries per liter, is variable
- 57 • WELL FROM WHICH WATER SAMPLE FOR IODINE-129 WAS COLLECTED IN 1990-91--Number, 57, is local well identifier; P indicates well obtains water from a perched ground-water zone. See table 2 for iodine-129 concentrations in specific wells

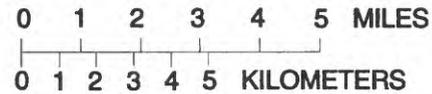
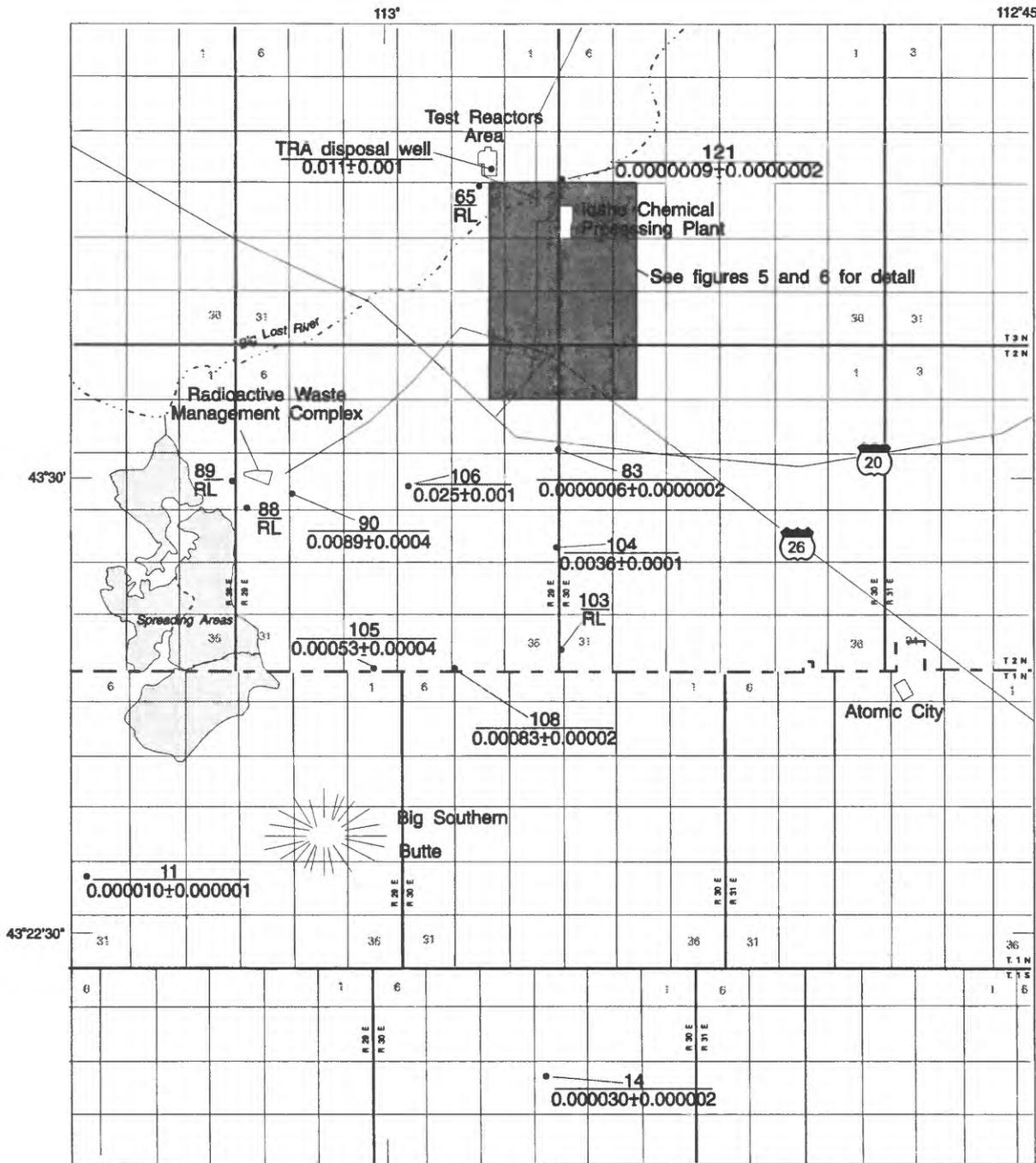
Figure 5.--Areal distribution of iodine-129 in the Snake River Plain aquifer near the Idaho Chemical Processing Plant, 1990-91.



EXPLANATION

- 1 — LINE OF EQUAL IODINE-129 CONCENTRATION--Interval, in picocuries per liter, is variable
-  APPROXIMATE LIMIT OF AREA IN WHICH IODINE-129 CONCENTRATION EXCEEDS 0.25 PICOCURIE PER LITER
- 77 • WELL FROM WHICH WATER SAMPLE FOR IODINE-129 WAS COLLECTED IN 1986--Number, 77, is local well identifier. See table 2 for iodine-129 concentrations in specific wells

Figure 6.--Areal distribution of iodine-129 in the Snake River Plain aquifer near the Idaho Chemical Processing Plant, 1986 (Modified from Mann and others, 1988).



EXPLANATION

- $\frac{106}{0.025 \pm 0.001}$ WELL--Upper entry, 106, is the well identifier; lower entry, 0.025±0.001, is the iodine-129 concentration and analytical uncertainty in picocuries per liter. RL indicates the concentration of iodine-129 was less than the 3σ reporting level
- BOUNDARY OF THE IDAHO NATIONAL ENGINEERING LABORATORY

Figure 7.--Distribution of iodine-129 in the Snake River Plain aquifer in areas adjacent to the Idaho Chemical Processing Plant, 1990-91.

River. Therefore, there is a 95-percent probability that the calculated background concentration is less than or equal to 0.0000012 pCi/L —the concentration plus $1.645s$ or $0.0000009+0.0000003 \text{ pCi/L}$ —and concentrations greater than 0.0000012 pCi/L result from iodine-129 in wastewater at the ICPP. Water samples with concentrations less than the reporting level (table 2) cannot be compared to the background concentration. The only water sample that contained a concentration less than the calculated background concentration was from well 83, in which the concentration was $0.0000006\pm 0.0000002 \text{ pCi/L}$ (fig. 7). Water from wells 105 and 108, at the southern boundary of the INEL, and wells 11 and 14, south of the INEL about 4 and 8 mi (table 2), respectively, contained small concentrations of iodine-129 that is attributed to wastewater disposal at the ICPP; the concentrations are about one to two orders of magnitude greater than the calculated background concentration.

Vertical Distribution of Iodine-129 in the Snake River Plain Aquifer

Iodine-129 is not distributed uniformly with depth in the Snake River Plain aquifer near the ICPP. For example, wells 51 and 123 are about 590 ft apart (fig. 5), but they are open to different intervals of the aquifer. Because of their proximity to one another, if they were open to the same intervals and iodine-129 were distributed uniformly with depth, the concentrations of iodine-129 in water samples from the two wells should be comparable. Well 51 obtains water from the interval between 475 and 659 ft below land surface (table 1) and water from the well contained $0.28\pm 0.01 \text{ pCi/L}$ of iodine-129 (table 2). Well 123, however, obtains water from the interval between 461 and 475 ft below land surface, and the water contained $1.00\pm 0.05 \text{ pCi/L}$.

Water samples from wells 34, 35, and 36 also show that the concentrations of iodine-129 decrease with depth. These wells are southwest of the ICPP and are about 500 ft apart; well 34 is about midway between wells 35 and 36 (fig. 5). Wells 35 and 36 obtain water from between 472 and 578 ft, and 471 and 567 ft below land surface,

respectively, and well 34 obtains water from the interval between 499 and 700 ft. The concentration of iodine-129 in water from well 34 was $0.39\pm 0.02 \text{ pCi/L}$ and was less than the concentration of $0.57\pm 0.03 \text{ pCi/L}$ at well 35 and $1.17\pm 0.05 \text{ pCi/L}$ at well 36; replicate water samples from well 36 contained 1.19 ± 0.03 and $1.74\pm 0.06 \text{ pCi/L}$, respectively (table 2).

A similar comparison for wells 59 and 122 shows that concentrations of iodine-129 increase with depth in the aquifer. Wells 59 and 122 are 260 ft apart and obtain water from intervals between 464 and 657 ft, and 456 and 475 ft below land surface, respectively. The concentration of iodine-129 in water from well 122 was $0.37\pm 0.02 \text{ pCi/L}$ and was less than that for well 59— $0.57\pm 0.02 \text{ pCi/L}$.

Several factors affect the vertical distribution of iodine-129 in the aquifer:

- (1) The horizontal and vertical hydraulic conductivities of the basaltic lava flows differ from flow to flow and within a specific flow;
- (2) the concentration of iodine-129 in wastewater at the ICPP changed from year to year (fig. 2);
- (3) the ICPP disposal well failed three times between 1952—when it was drilled and constructed—and 1984—when it was no longer routinely used to dispose of wastewater;
- (4) the amount of recharge to the aquifer varies; and
- (5) vertical hydraulic gradients in the aquifer are not uniform owing to changes in horizontal and vertical hydraulic conductivities.

Because of the complex nature of the aquifer, horizontal hydraulic conductivities differ by nearly six orders of magnitude. Horizontal hydraulic conductivity estimated by Ackerman (1991, p. 30) ranged from 8.6×10^{-3} to $5.5\times 10^3 \text{ ft/day}$ for wells at and near the INEL. The vertical hydraulic conductivity of the aquifer may have a comparable

range depending on the lenticularity and density of basalt flows, the degree to which the flows are fractured vertically, and whether sedimentary interbeds are present between individual flows or groups of flows. Unfractured and dense zones in the basalt flows, and silt and clay in sedimentary interbeds impede the vertical movement of water, whereas vertical fractures and coarse-grained sediment more readily transmit water vertically. The stratigraphy of the unsaturated zone and the upper part of the Snake River Plain aquifer at the ICPP are described by Anderson (1991).

The ICPP disposal well originally was drilled to a depth of 598 ft and the casing was perforated in the intervals between 412 and 452 ft, and 490 and 593 ft below land surface (table 1). Geophysical logs of June 1970 indicated that the casing was corroded and broken or missing in places. In January 1971 the well was cleaned out to 596 ft and new casing was installed and perforated between 508 and 588 ft. In July 1982, a caliper log showed that there were separations in the casing installed in January 1971 and that the casing had collapsed at about 435 ft. The well was cleaned out to 584 ft, and new casing liner was installed and perforated between 470 and 510 ft; the casing installed in January 1971 was still in place and the perforations were open between 510 and at least 560 ft. In 1988, however, a television log showed that the well was filled in to about 480 ft. Given the different intervals open to the aquifer and the unsaturated zone above the water table—about 450 ft below land surface—and the unknown rate at which the well filled with sediment, the zones into which the wastewater was injected changed from time to time. Because it is likely that sediment progressively accumulated in the bottom of the well, a large part of the wastewater probably was injected in the interval between 412 and 550 ft and could have been injected in the interval between 412 and 480 ft.

A water sample from well 50, which obtains water from a zone perched about 50 ft above the water table for the Snake River Plain aquifer, contained 1.20 ± 0.04 pCi/L of iodine-129 (table 2); well 50 is about 350 ft north of the ICPP disposal well (fig. 5). The source of the water in well 50 and the extent of the perched ground-water zone

that well 50 penetrates are not fully known; however, water-level changes in well 50 from 1965 to 1971 indicate that at least part of the water in the perched ground-water zone came from the ICPP disposal well. In October 1966, the depth to water in well 50 was 377 ft below land surface (fig. 8). In July 1968, the depth to water was 295 ft below land surface—a rise in water level of 82 ft. From July 1968 to March 1971, the water level ranged from 287 to 322 ft below land surface. In April 1971, during or after rehabilitation of the ICPP disposal well, the depth to water in well 50 declined to 360 ft below land surface and has ranged from 371 to 382 ft since then. The rise in water level in 1966–68 indicates that casing in the ICPP disposal well failed during that time. The decline in water level in April 1971 demonstrates that the disposal well was the source of at least part of the water in the perched ground-water zone from 1966–68 to 1971. Whether the iodine-129 and other wastewater constituents in water from well 50 are totally from the disposal well, or are also from other sources cannot be ascertained. Wastewater was injected into well 50 during August and September 1971, and from December 1971 to February 1972. The wastewater source and contaminants that may have been in the wastewater are unknown.

The depth to water in well 50 did not change appreciably in 1984 when routine use of the ICPP disposal well was discontinued (fig. 8). This implies that the disposal well was not the only source of the water in the perched ground-water zone penetrated by well 50. The general rise in the ground-water level since 1987 indicates that the perched ground-water zone is being recharged. The source or sources of the recharge are unknown. Data provided by Cecil and others (1991) show that the tritium and strontium-90 concentrations in water from well 50 are markedly greater than those in water from six wells that are open to a shallower perched ground-water zone that underlies the ICPP disposal ponds. The chloride concentration in water from well 50, however, is less than that in water from the six wells near the disposal ponds. These data indicate that the source of water at well 50 is not the disposal ponds. The perched ground-water zone penetrated by well 50 is, nevertheless, a potential

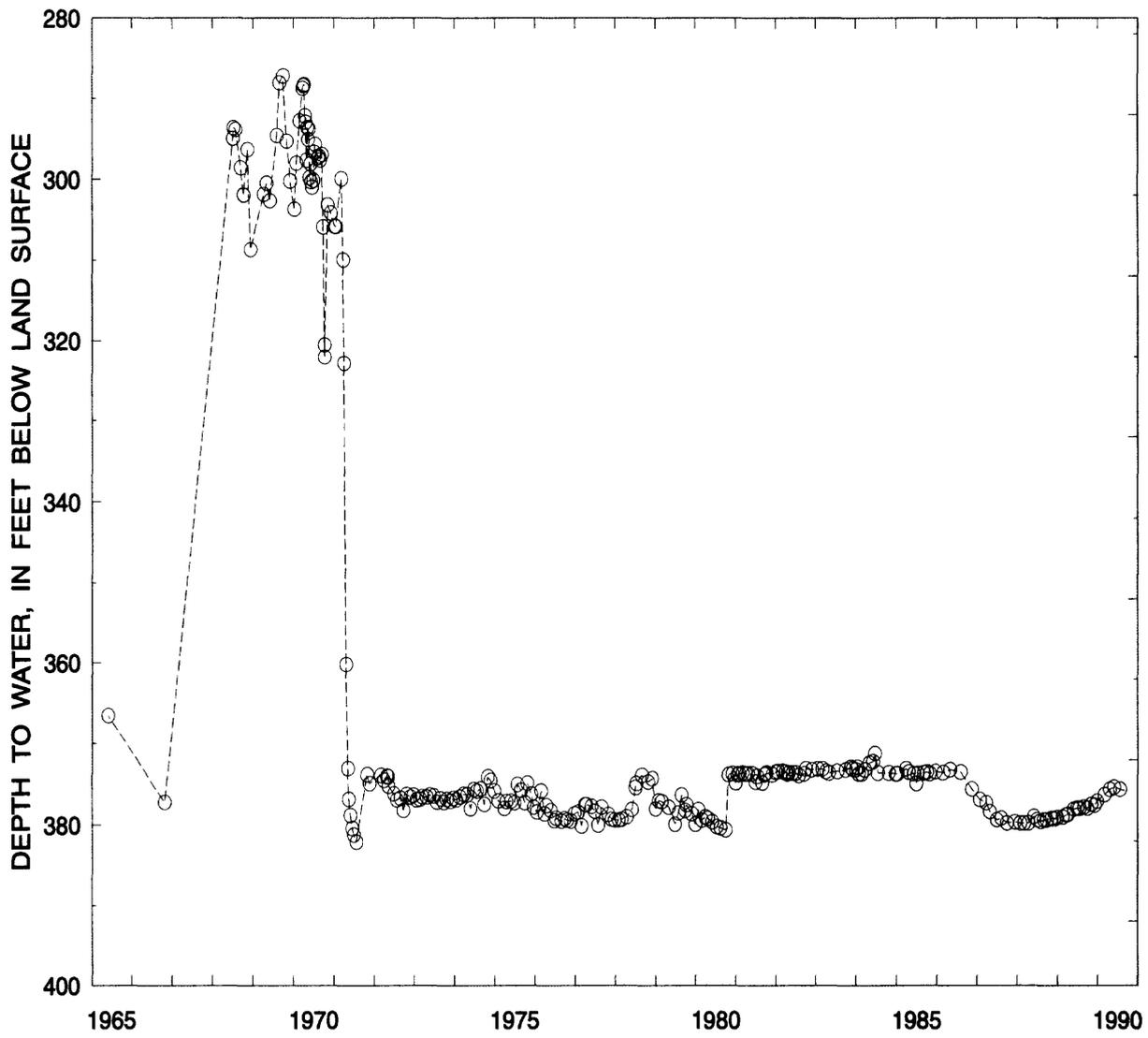


Figure 8.--Depth to water in well 50 at the Idaho Chemical Processing Plant.

source of iodine-129 in the Snake River Plain aquifer.

Recharge from the infiltration of streamflow in the Big Lost River and from the disposal ponds routinely used to dispose of wastewater beginning in February 1984 also affects the vertical distribution of iodine-129 in the aquifer. Recharge from the river and disposal ponds dilutes the concentrations in the aquifer. Near the river and disposal ponds, iodine-129 concentrations in the aquifer increase with depth, as indicated by the concentrations in water from wells 59 and 122 as described previously.

In 1990 and 1991, a heat-pulse flowmeter log indicated that water in the uncased intervals of wells 44, 45, and 46 was moving upward through the well bore. The upward movement demonstrates that there is an upward vertical hydraulic gradient in the aquifer that would prevent wastewater injected into the upper part of the aquifer from circulating to lower parts. The upward vertical gradient supports conclusions by Robertson and others (1974) that waste plumes in the aquifer generally remain in the uppermost 250 ft and that this part of the aquifer is somewhat separated from and is more permeable than deeper parts. The fact that tritium is not detected in water from some wells open to the uppermost 200 to 500 ft of the aquifer (Mann and Cecil, 1990, p. 18) also support the conclusions by Robertson and others (1974). However, from 1953 to 1984, when wastewater was injected into the aquifer, the vertical hydraulic gradient in the vicinity of the ICPP disposal well probably was downward owing to the injection process.

Iodine-129 Movement and Ground-Water Flow Velocities in the Snake River Plain Aquifer

The movement of iodine-129 in the Snake River Plain aquifer can be used to calculate ground-water flow velocities. Velocities of ground-water flow in the aquifer at the INEL previously have been estimated on the basis of tracer studies and the first arrivals of tritium. Tracer studies indicate that ground-water-flow velocities range from 5 to 20 ft/day with an average

near 10 ft/day (Robertson and others, 1974, p. 13). Estimates of ground-water-flow velocities based on the apparent velocity of tritium migration range from 4 to 5 ft/day (Barraclough and others, 1982, p. 43, and Pittman and others, 1988, p. 51).

Given that the iodine-129 concentrations in water from wells 11 and 14 are about one order of magnitude greater than the assumed background concentration, a minimum ground-water velocity can be calculated using the distance between the ICPP disposal well and wells 11 and 14, and the maximum known time it took for iodine-129 to travel that distance. The estimated distances along ground-water flowpaths between the disposal well and wells 11 and 14 are about 79,000 and 86,000 ft, respectively. Wastewater disposal at the ICPP began in 1953 and iodine-129 was detected in water from wells 11 and 14 in 1991. Velocities calculated by dividing the distance by the time of travel are 5.7 ft/day for well 11 and 6.2 ft/day for well 14; actual velocities could be greater because first arrival times are not known. However, it is reasonable to assume that the average velocity is at least 6 ft/day.

The average ground-water velocity of at least 6 ft/day calculated using iodine-129 data is slightly greater than the range of 4 to 5 ft/day previously estimated by Barraclough and others (1982, p. 43) and Pittman and others (1988, p. 51). However, it is markedly less than the average of 10 ft/day estimated by Robertson and others (1974, p. 13). Because of the comparatively short 12.3-year half-life of tritium, the comparatively large method detection limit of 500 pCi/L typically used for tritium analyses, and dispersion and dilution of the tritiated water in the aquifer, the velocities calculated using tritium arrivals are minimal values. Therefore, the average velocity of at least 6 ft/day calculated using the iodine-129 data may more accurately represent the lower bound of ground-water-flow velocities in the aquifer between the ICPP and wells 11 and 14.

Comparison of Iodine-129 Concentrations to Drinking-Water Standards

The proposed maximum contaminant level for iodine-129 in drinking water for community and

non-transient, non-community public water systems is 21 pCi/L. The concentration of 21 pCi/L corresponds to a 4 millirem per year effective dose equivalent and assumes a lifetime intake of 2 L/day of water (U.S. Environmental Protection Agency, 1991).

In 1990–91, the concentrations of iodine-129 in water from wells in the Snake River Plain aquifer were less than the proposed maximum contaminant level of 21 pCi/L. The largest concentration of iodine-129 was 3.82 ± 0.19 pCi/L in water from well 42—a ground-water quality monitoring well near the ICPP (see table 2 and fig. 5). The largest concentration of iodine-129 in water from a well used for drinking-water was 0.28 ± 0.02 pCi/L in well CFA-1 (table 2).

SUMMARY

Iodine-129 is one of the fission products contained in wastewater at the ICPP. Most of the wastewater was injected directly to the Snake River Plain aquifer through a 598-ft-deep disposal well or was discharged to unlined disposal ponds. Because of its 15.7 million-year half-life, iodine-129 is a permanent environmental pollutant. In 1990–91, water samples were collected from 51 wells that obtain water from the aquifer and analyzed for iodine-129. The water samples were analyzed using the AMS method. The detection limit for this method is from two to six orders of magnitude more sensitive than the detection limit for neutron-activation method, which was used for samples collected in 1977, 1981, and 1986. Because of the more sensitive method detection limit, iodine-129 was detected in water from a larger number of wells distributed over a larger area.

Between 1953 and 1990, an estimated 0.56 to 1.18 Ci of iodine-129 were contained in wastewater discharged to the disposal well and ponds at the ICPP. In general, concentrations of iodine-129 in wastewater decreased between 1978 and 1990. In 1990–91, concentrations in water from the Snake River plain aquifer at and near the INEL ranged from 0.0000006 ± 0.0000002 to 3.82 ± 0.19 pCi/L; for the purpose of comparison, the proposed maximum contaminant level for

iodine-129 in drinking water is 21 pCi/L. The mean concentration of iodine-129 in water from 18 wells was 0.81 ± 0.19 pCi/L in 1990–91 compared with 1.30 ± 0.26 pCi/L in 1986. Concentrations of iodine-129 in the ground water were greater than 0.25 pCi/L in a 3.4-mi² area; in 1986 concentrations were greater than 0.25 pCi/L in a 4.3-mi² area.

Iodine-129 is not distributed uniformly with depth in the aquifer. Water samples from five wells that are south and southwest of the disposal well and completed at different intervals below the water table indicate that concentrations of iodine-129 decrease with depth at some locations. However, water samples from two wells southeast of the disposal well indicate that concentrations also increase with depth at some locations.

Velocities of ground-water flow in the Snake River Plain aquifer at the INEL previously have been estimated on the basis of tracer studies and the first arrivals of tritium. Tracer studies indicate that flow velocities average about 10 ft/day; ground-water-flow velocities based on the apparent velocity of tritium migration ranges from 4 to 5 ft/day. Velocities calculated using iodine-129 concentrations in water from wells about 15 to 16 mi hydraulically downgradient from the ICPP are at least 6 ft/day.

REFERENCES CITED

- Ackerman, D.J., 1991, Transmissivity of the Snake River Plain aquifer at the Idaho National Engineering Laboratory, Idaho: U.S. Geological Survey Water-Resources Investigations Report 91-4058, 35 p.
- Anderson, S.R., 1991, Stratigraphy of the unsaturated zone and uppermost part of the Snake River Plain aquifer at the Idaho Chemical Processing Plant and Test Reactors Area, Idaho National Engineering Laboratory, Idaho: U.S. Geological Survey Water-Resources Investigations Report 91-4010, 71 p.
- Barracough, J.T., and Jensen, R.G., 1976, Hydrologic data for the Idaho National Engineering Laboratory site, Idaho, 1971 to 1973: U.S. Geological Survey Open-File Report 75-318, 52 p.

- Barraclough, J.T., Lewis, B.D., and Jensen, R.G., 1982, Hydrologic conditions at the Idaho National Engineering Laboratory, Idaho—emphasis, 1974-1978: U.S. Geological Survey Water-Supply Paper 2191, 52 p.
- Bennett, C.M., 1990, Streamflow losses and ground-water level changes along the Big Lost River at the Idaho National Engineering Laboratory, Idaho: U.S. Geological Survey Water-Resources Investigations Report 90-4067, 49 p.
- Cecil, L.D., Orr, B.R., Norton, Teddy, and Anderson, S.R., 1991, Formation of perched ground-water zones and concentrations of selected chemical constituents in water, Idaho National Engineering Laboratory, Idaho, 1986-88: U.S. Geological Survey Water-Resources Investigations Report 91-4166, 53 p.
- Claassen, H.C., 1982, Guidelines and techniques for obtaining water samples that accurately represent the water chemistry of an aquifer: U.S. Geological Survey Open-File Report 82-1024, 49 p.
- Currie, L.A., 1968, Limits for qualitative detection and quantitative determination—application to radiochemistry: *Analytical Chemistry*, v. 40, no. 3, p. 586-593.
- EG&G Idaho, Inc., 1979, Radioactive waste management information for 1978: U.S. Department of Energy Report IDO-10055(78), 291 p.
- 1980, Radioactive waste management information for 1979: U.S. Department of Energy Report IDO-10055(79), 284 p.
- 1981, Radioactive waste management information for 1980: U.S. Department of Energy Report IDO-10055(80), 273 p.
- 1982, Radioactive waste management information for 1981: U.S. Department of Energy Report IDO-10055(81), 270 p.
- 1983, Radioactive waste management information for 1982: U.S. Department of Energy Report IDO-10055(82), 276 p.
- 1984, Radioactive waste management information for 1983: U.S. Department of Energy Report DOE/ID-10055(83), 272 p.
- 1985, Radioactive waste management information for 1984: U.S. Department of Energy Report DOE/ID-10055(84), 275 p.
- 1986, Radioactive waste management information for 1985: U.S. Department of Energy Report DOE/ID-10055(85), 310 p.
- 1987, Radioactive waste management information for 1986: U.S. Department of Energy Report DOE/ID-10055(86), 388 p.
- 1988, Radioactive waste management information for 1987 and Record-to-Date: U.S. Department of Energy Report DOE/ID-10054(87), [variously paged].
- 1989, Radioactive waste management information for 1988 and Record-to-Date: U.S. Department of Energy Report DOE/ID-10054(88), [variously paged].
- 1990, Radioactive waste management information for 1989 and Record-to-Date: U.S. Department of Energy Report DOE/ID-10054(89), [variously paged].
- 1991, Radioactive waste management information for 1990 and Record-to-Date: U.S. Department of Energy Report DOE/ID-10054(90), [variously paged].
- Elmore, David, and Phillips, F.M., 1987, Accelerator mass spectrometry for measurement of long-lived radioisotopes: *Science*, v. 236, p. 543-550
- Fabryka-Martin, J., Bentley, H., Elmore, D., and Airey, P.L., 1985, Natural iodine-129 as an environmental tracer: *Geochimica et Cosmochimica Acta*, Vol. 49, p. 337-347.
- Furman, N.H., ed., 1962, Standard methods of chemical analysis, (6th ed.), v. 1, New York, D. Van Nostrand Co, 1401 p.
- Harenberg, W.A., Jones, M.L., O'Dell, I., and Cordes, S.C., 1989, Water-resources data, Idaho, water year 1988: U.S. Geological Survey Water-Data Report ID-88-1, 669 p.
- 1990, Water-resources data, Idaho, water year 1989: U.S. Geological Survey Water-Data Report ID-89-1, 681 p.
- Harenberg, W.A., Jones, M.L., O'Dell, I., Brennan, T.S., and Lehmann, A.K., 1991, Water-resources data, Idaho, water year 1990: U.S. Geological Survey Water-Data Report ID-90-1, 641 p.
- Kieser, L., ed., 1989, 1989 Annual Report, IsoTrace Laboratory: Toronto, University of Toronto, Ontario, Canada, 51 p.

- Kilius, L.R., 1990, 1990 Annual report, IsoTrace Laboratory, Toronto, University of Toronto, Ontario, Canada, 50 p.
- Kilius, L.R., Rucklidge, J.C., and Litherland, A.E., 1987, Accelerator mass spectrometry of ^{129}I at IsoTrace, 1987: in Gove, H.E., Litherland, A.E., and Elmore, David, eds., Accelerator mass spectrometry: International Symposium on Accelerator Mass Spectrometry, 4th, Niagara-on-the-Lake, Ontario, Canada, April 27-30, 1987 [Proceedings], p. 72-76.
- Kleinberg, Jacob. and Cowan, G.A., 1960, The radiochemistry of fluorine, chlorine, bromine, and iodine: [available from National Technical Information Service, Springfield, VA 22161 as NAS-NS 3005].
- Kubik, P.W., Elmore, David, Hemmick, T.K., Gove, H.F., Fehn, Udo, Teng, T.T.D., Jiang, Songshing, and Pullai, Sharrin, 1987, Accelerator mass spectrometry at the University of Rochester, in Gove, H.E., Litherland, A.E., and Elmore, David, eds., Accelerator mass spectrometry: International Symposium on Accelerator Mass Spectrometry, 4th, Niagara-on-the-Lake, Ontario, Canada, April 27-30, 1987 [Proceedings], p. 138-142.
- Lewis, B.D., and Jensen, R.G., 1985, Hydrologic conditions at the Idaho National Engineering Laboratory, Idaho--1979-1981 update: U.S. Geological Survey Hydrologic Investigations Atlas HA-674, 2 sheets.
- Mann, L.J., and Cecil, L.D., 1990, Tritium in ground water at the Idaho National Engineering Laboratory, Idaho: U.S. Geological Survey Water-Resources Investigations Report 90-4090, 35 p.
- Mann, L.J., Chew, E.W., Morton, J.S., and Randolph, R.B., 1988, Iodine-129 in the Snake River Plain aquifer at the Idaho National Engineering Laboratory, Idaho: U.S. Geological Survey Water-Resources Investigations Report 88-4165, 27 p.
- National Council on Radiation Protection and Measurements, 1983, Iodine-129--Evaluation of releases from nuclear power generation: National Council on Radiation Protection and Measurements Report No. 75, 74 p.
- Orr, B.R., and Cecil, L.D., 1991, Hydrologic conditions and distribution of selected chemical constituents in water, Snake River Plain aquifer, Idaho National Engineering Laboratory, Idaho, 1986 to 1988: U.S. Geological Survey Water-Resources Investigations Report 91-4047, 56 p.
- Pittman, J.R., Jensen, R.G., and Fischer, P.R., 1988, Hydrologic conditions at the Idaho National Engineering Laboratory, 1982 to 1985: U.S. Geological Survey Water-Resources Investigations Report 89-4008, 73 p.
- Robertson, J.B., Schoen, Robert, and Barraclough, J.T., 1974, The influence of liquid waste disposal on the geochemistry of water at the National Reactor Testing Station, Idaho: U.S. Geological Survey Open-File Report, IDO-22053, 231 p.
- Suter, M., 1990, Accelerator mass spectrometry—state of the art in 1990: Nuclear Instruments and Methods in Physics Research, B52, p. 211-223.
- U.S. Environmental Protection Agency, 1991, Proposed drinking water limits for radionuclides: U.S. Federal Register, v. 56, no. 138, p. 33,050-33,127.
- White, S.S., 1977, Radioactive waste management information for 1976: Energy Research and Development Administration Report IDO-10055(76), 287 p.
- 1978, Radioactive waste management information for 1977: U.S. Department of Energy Report IDO-10055(77), 289 p.
- Wood, W.W., 1981, Guidelines for collection and field analysis of ground-water samples for selected unstable constituents: U.S. Geological Survey Techniques of Water-Resources Investigations, book 1, chap. D2, 24 p.