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Iodine-129 in the Eastern Snake River Plain Aquifer at and near the Idaho National Laboratory, Idaho, 2010–12



Scientific Investigations Report 2013–5195

U.S. Department of the Interior U.S. Geological Survey

Cover: U.S. Geological Survey scientist collecting Iodine-129 sample at Kimama water supply well, Kimama, Idaho, October 6, 2010. (Photograph courtesy of Linda Davis, U.S. Geological Survey, Idaho National Laboratory Project Office.)

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By Roy C. Bartholomay

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U.S. Geological Survey

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Conversion Factors, Datums, and Abbreviations and Acronyms

Conversion Factors

Inch/Pound to SI

Multiply	Ву	To obtain
	Length	
foot (ft)	0.3048	meter (m)
mile (mi)	1.609	kilometer (km)
square mile (mi ²)	2.590	square kilometer (km ²)
	Area	
acre	4,047	square meter (m ²)
	Flow rate	
acre-foot (acre-ft)	1,233	cubic meter (m ³)
foot per mile (ft/mi)	0.1894	meter per kilometer (m/km)
foot per second (ft/s)	0.3048	meter per second (m/s)
foot per day (ft/d)	0.3048	meter per day (m/d)
cubic foot per second (ft ³ /s)	0.02832	cubic meter per second (m^3/s)
gallon per minute (gal/min)	0.06309	liter per second (L/s)
	Radioactivity	
picocurie per liter (pCi/L)	0.037	becquerel per liter (Bq/L)
attocurie per liter (aCi/L)	3.7×10 ⁻⁸	becquerel per liter (Bq/L)
	Transmissivity*	
foot squared per day (ft ² /d)	0.0929	meter squared per day (m ² /d)

SI to Inch/Pound

Multiply	Ву	To obtain
	Volume	
liter (L)	33.82	ounce, fluid (fl. oz)
liter (L)	2.113	pint (pt)
liter (L)	1.057	quart (qt)
liter (L) 0.2642 gallon (gal)		gallon (gal)
	Mass	
gram (g)	0.03527	ounce, avoirdupois (oz)

*Transmissivity: The standard unit for transmissivity is cubic foot per day per square foot times foot of aquifer thickness [(ft³/d)/ft²]ft. In this report, the mathematically reduced form, foot squared per day (ft²/d), is used for convenience.

Concentrations of chemical constituents in water are given either in milligrams per liter (mg/L) or micrograms per liter (μ g/L).

Conversion Factors, Datums, and Abbreviations and Acronyms

Datums

Vertical coordinate information is referenced to the National Geodetic Vertical Datum of 1929 (NGVD 29).

Horizontal coordinate information is referenced to the North American Datum of 1927 (NAD 27).

Altitude, as used in this report, refers to distance above the vertical datum.

Abbreviations and Acronyms

Abbreviation or acronym	Definition					
129	lodine-129					
AMS	accelerator mass spectrometer					
ATRC	Advanced Test Reactor Complex					
CFA	Central Facilities Area					
Ci	curie					
DOE	U.S. Department of Energy					
ESRP	eastern Snake River Plain					
INL	Idaho National Laboratory					
INTEC	Idaho Nuclear Technology and Engineering Center					
MCL	maximum contaminant level					
NRF	Naval Reactors Facility					
RTC	Reactor Technology Complex					
RWMC	Radioactive Waste Management Complex					
PRIME	Purdue Rare Isotope Measurement					
TRA	Test Reactor Area					
USEPA	U.S. Environmental Protection Agency					
USGS	U.S. Geological Survey					

Iodine-129 in the Eastern Snake River Plain Aquifer at and near the Idaho National Laboratory, Idaho, 2010–12

By Roy C. Bartholomay

Abstract

From 1953 to 1988, approximately 0.941 curies of iodine-129 (¹²⁹I) were contained in wastewater generated at the Idaho National Laboratory (INL) with almost all of this wastewater discharged at or near the Idaho Nuclear Technology and Engineering Center (INTEC). Most of the wastewater containing ¹²⁹I was discharged directly into the eastern Snake River Plain (ESRP) aquifer through a deep disposal well until 1984; lesser quantities also were discharged into unlined infiltration ponds or leaked from distribution systems below the INTEC.

During 2010-12, the U.S. Geological Survey in cooperation with the U.S. Department of Energy collected groundwater samples for ¹²⁹I from 62 wells in the ESRP aquifer to track concentration trends and changes for the carcinogenic radionuclide that has a 15.7 million-year half-life. Concentrations of ¹²⁹I in the aquifer ranged from 0.0000013±0.0000005 to 1.02±0.04 picocuries per liter (pCi/L), and generally decreased in wells near the INTEC, relative to previous sampling events. The average concentration of ¹²⁹I in groundwater from 15 wells sampled during four different sample periods decreased from 1.15 pCi/L in 1990-91 to 0.173 pCi/L in 2011-12. All but two wells within a 3-mile radius of the INTEC showed decreases in concentration, and all but one sample had concentrations less than the U.S. Environmental Protection Agency maximum contaminant level of 1 pCi/L. These decreases are attributed to the discontinuation of disposal of ¹²⁹I in wastewater and to dilution and dispersion in the aquifer. The decreases in ¹²⁹I concentrations, in areas around INTEC where concentrations increased between 2003 and 2007, were attributed to less recharge near INTEC either from less flow in the Big Lost River or from less local snowmelt and anthropogenic sources.

Although wells near INTEC sampled in 2011–12 showed decreases in ¹²⁹I concentrations compared with previously collected data, some wells south and east of the Central Facilities Area, near the site boundary, and south of the INL showed small increases. These slight increases are attributed to variable discharge rates of wastewater that eventually moved to these well locations as a pulse of water from a particular disposal period.

Wells sampled for the first time around the Naval Reactors Facility had ¹²⁹I concentrations slightly greater than background concentrations in the ESRP aquifer. These concentrations are attributed to either seepage of unknown wastewater sources discharged at the Naval Reactors Facility or seepage from air emission deposits from INTEC, or both.

In 2012, the U.S. Geological Survey collected discrete groundwater samples from 25 zones in 11 wells equipped with multilevel monitoring systems to help define the vertical distribution of ¹²⁹I in the aquifer. Concentrations ranged from 0.000006±0.000004 to 0.082±0.003 pCi/L. Two new wells completed in 2012 showed variability of up to one order of magnitude of concentrations of ¹²⁹I among various zones. Two other wells showed similar concentrations of ¹²⁹I in all three zones sampled. Concentrations were well less than the maximum contaminant level in all zones.

Introduction

The Idaho National Laboratory (INL), encompassing about 890 mi² of the eastern Snake River Plain (ESRP) in southeastern Idaho (fig. 1), is operated by the U.S. Department of Energy (DOE). The INL was established in 1949 for the development of peacetime atomic energy applications, nuclear safety research, defense programs, environmental research, and advanced energy concepts. Until 1993, uranium from spent nuclear fuel elements from government-owned reactors was recovered after reprocessing at the Idaho Nuclear Technology and Engineering Center (INTEC; fig. 1). As part of the fuel reprocessing activities, fission products were released in wastewater at the INTEC. Iodine-129 (129I), produced by the fission of uranium-235 and plutonium-239, was one of the products released in wastewater. Prior to 1984, most of the wastewater generated at the INTEC was injected directly to the ESRP 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 unlined infiltration ponds south of INTEC, which allow the wastewater to percolate through about 450 ft of basalt and sediment to the aquifer.



Figure 1. Location of the Idaho National Laboratory and selected wells, Idaho.

Iodine-129 in the ESRP aquifer originates from atmospheric deposition, rock weathering, and wastewater disposal (Mann and Beasley, 1994a). The amount of ¹²⁹I in the aquifer from atmospheric deposition and rock weathering is considered small, and is included in the estimated background concentration of 0.0000054 picocuries per liter (pCi/L) in the ESRP aquifer in eastern Idaho (Cecil and others, 2003). Mann and Beasley (1994a) reported that wastewater discharged to the injection well and infiltration ponds at the INTEC between 1953 and 1990 contained an estimated 0.56-1.18 curies (Ci) of ¹²⁹I. A more detailed estimate of wastewater discharge was performed by the DOE Idaho Operations Office (U.S. Department of Energy, 2004, appendix D), and results indicated that a maximum of 0.86 Ci of 129I was discharged to the aquifer through the injection well. In addition, about 0.08 Ci of ¹²⁹I were discharged to the infiltration ponds from 1984 to 1988 (Litteer, 1988; Mann and others, 1988, table 2; Litteer and Reagan, 1989), and about 0.001 Ci of ¹²⁹I was released at the INTEC Tank Farm between 1958 and 1986 (Cahn and others, 2006, table 5-2). Thus, the revised total released to the ESRP aquifer is about 0.941 Ci from INTEC. Therefore, some ¹²⁹I may still be present in perched zones around the INTEC. Some 129I also was discharged into the radioactive waste ponds at the Advanced Test Reactor Complex (ATRC), but annual concentrations of the discharge water generally were much less than 1 pCi/L; for example, EG&G Idaho, Inc. (1979) showed an average annual concentration of 0.0000128 pCi/L in the 1978 discharge water. Because of its 15.7 million-year half-life, ¹²⁹I released to the environment is a permanent addition to the global inventory (Mann and Beasley, 1994a).

Iodine-129 is a carcinogen, and communities downgradient of the INL may be concerned that ¹²⁹I disposed at the INL could be a health hazard. The current (1976) U.S. Environmental Protection Agency maximum contaminant level (MCL) for ¹²⁹I in drinking water is 1 pCi/L (U.S. Environmental Protection Agency, 1976, appendix B). The MCL is based on the average concentration in public drinking water supplies that will yield an annual whole-body dose equivalent to about 4 millirem for man-made betaparticle and photon-emitting radionuclides; the proposed MCL based on effective dose equivalent for ¹²⁹I is 21 pCi/L (U.S. Environmental Protection Agency, 2000). To evaluate the potential hazards, the U.S. Geological Survey (USGS) in cooperation with the DOE, have periodically monitored for ¹²⁹I in groundwater from the ESRP aquifer at and downgradient of the INTEC since 1977. Monitoring programs from 1977, 1981, 1986, 1990–91, 2003, and 2007 were summarized by Mann and others (1988); Mann and Beasley (1994b); and Bartholomay (2009). This report summarizes concentrations in the ESRP aquifer from 2010–12.

Purpose and Scope

The USGS collected water samples from wells during 2010 through 2012 to evaluate recent concentrations of ¹²⁹I in the ESRP aquifer. This report describes the results of water samples collected for ¹²⁹I analyses and shows the comparisons with concentrations from previously collected samples, and background levels for wells that were sampled for the first time.

In October 2010, two samples were collected from two wells at Kimama, Idaho (fig. 1) to determine whether water discharged at the INL had reached the central part of the ESRP aquifer downgradient of the INL. In October and November 2011, 31 samples were collected from 29 wells that are used to monitor the ESRP aquifer at or near the INL. In 2012, 22 samples were collected from an additional 20 wells that are used to monitor the ESRP aquifer. In addition, 25 zones in 11 wells equipped with multi-level monitoring systems (MLMS) (figs. 2 and 3) were sampled in 2012. Six replicates, a source solution blank, and an equipment blank were collected for quality control purposes. The wells were sampled to determine the current ¹²⁹I concentrations in the ESRP aquifer. Samples were analyzed using the Accelerator Mass Spectrometer (AMS) method at the Purdue Rare Isotope Measurement (PRIME) laboratory, Purdue University, West Lafayette, Indiana, so concentrations could be compared with past data to determine concentration changes.

Geohydrologic Setting

The INL is located on the west-central part of the ESRP. The ESRP is a northeast-trending structural basin about 200 mi long and 50–70 mi wide (fig. 1). The basin, bounded by faults on the northwest and by downwarping and faulting on the southeast, has been filled with basaltic lava flows interbedded with terrestrial sediments. The basaltic rocks and sedimentary deposits combine to form the ESRP aquifer, which is the main source of groundwater on the plain.



Figure 2. Location of wells at and near the Idaho National Laboratory, Idaho.

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Base from U.S. Geological Survey digital data, 1:24,000 and 1:100,000 Universal Transverse Mercator projection, Zone 12 Datum is North American Datum of 1927

Figure 3. Location of wells at and near the Advanced Test Reactor Complex and Idaho Nuclear Technology and Engineering Center, Idaho National Laboratory, Idaho.

The ESRP aquifer is one of the most productive aquifers in the United States (U.S. Geological Survey, 1985, p. 193). Movement of water in the aquifer generally is from northeast to southwest, and water eventually discharges to springs along the Snake River downstream of Twin Falls, Idaho—about 100 mi southwest of the INL. Water moves horizontally through basalt interflow zones and vertically through joints and interfingering edges of interflow zones. Infiltration of surface water, heavy pumpage, geologic conditions, and seasonal fluxes of recharge and discharge locally affect the movement of groundwater (Garabedian, 1986). Recharge to the ESRP aquifer is primarily from infiltration of applied irrigation water, infiltration of streamflow, groundwater inflow from adjoining mountain drainage basins, and infiltration of precipitation.

At the INL, depth to water in wells completed in the ESRP aquifer ranges from about 200 ft in the northern part of the site to more than 900 ft in its southeastern part. A significant proportion of the groundwater moves through the upper 200–800 ft of basaltic rocks (Mann, 1986, p. 21). Ackerman (1991, p. 30) and Bartholomay and others (1997, table 3) reported a range of transmissivity of basalt in the upper part of the aquifer of 1.1–760,000 ft²/d. The hydraulic gradient at the INL ranges from 2 to 10 ft/mi, with an average of about 4 ft/mi (Davis, 2010, fig. 9). Horizontal flow velocities of 2-20 ft/d have been calculated based on the movement of various constituents in several areas of the aquifer at the INL (Robertson and others, 1974; Mann and Beasley, 1994b; Cecil and others, 2000; Busenberg and others, 2001). These flow rates equate to a travel time of about 70-700 years for water beneath the INL to travel to springs that discharge at the terminus of the ESRP aquifer near Twin Falls, Idaho (fig. 1). Localized tracer tests at the INL have shown vertical and horizontal transport rates as high as 60-150 ft/d (Nimmo and others, 2002; Duke and others, 2007).

Previous Investigations

Many investigations have been done to evaluate the geology and hydrology of the ESRP aquifer at the INL. A comprehensive listing of publications by the USGS at the INL is available at <u>http://id.water.usgs.gov/projects/INL/pubs.html</u> (U.S. Geological Survey, 2013a).

Previous investigations of ¹²⁹I in water from the ESRP aquifer include those by Barraclough and others (1982), Lewis and Jensen (1984), Mann and others (1988), Mann and Beasley (1994a; 1994b), Cecil and others (2003), U.S. Department of Energy (2004, 2007, 2008, 2012), Hall (2006), Forbes and others (2007), and Bartholomay (2009). Results from April 1977 sampling for ¹²⁹I in 13 wells indicated concentrations ranging from 0.9 to 27 pCi/L for statistically

positive values (Barraclough and others, 1982, fig. 42), and since discharge began in 1953, ¹²⁹I was identified in wells less than 3 mi from the disposal well. In October 1981, concentrations of ¹²⁹I ranged from 0.05 to 41 pCi/L (Lewis and Jensen, 1984) with statistically positive values present in 24 wells (Mann and others, 1988, table 1), and since discharge began in 1953, ¹²⁹I was identified in wells approximately 6.3 mi from the disposal well. Increased sensitivity because of an increase in sample volume from 1 to 4 L and improvement in analysis methods allowed a four-fold reduction in reporting level for ¹²⁹I analyses from 1977 to 1981 (Lewis and Jensen, 1985). This increase in sensitivity of analyses (Lewis and Jensen, 1985), along with a more extensive set of wells sampled (20 in 1977 and 32 in 1981) were factors contributing to the increase in the identified size of the ¹²⁹I plume for that time period. In August 1986, ¹²⁹I concentrations ranged from 0.49 ± 0.12 to 3.6 ± 0.4 pCi/L for 20 wells, with concentrations greater than the reporting level (Mann and others, 1988), and ¹²⁹I had migrated about the same distance from the disposal well as in 1981. The large decrease in the maximum concentration between 1981 and 1986 was attributed to changes in disposal practices at the INTEC, reduction in the mass of ¹²⁹I in wastewater, and increased dilution in the mid-1980s from a large amount of flow in the Big Lost River and increased recharge to the aquifer (Mann and others, 1988).

Prior to the 1990-91 data collection, neutron activation methods were used for analyses. During 1990-91, Mann and Beasley (1994b) collected samples from 51 wells at and near the INL, and they analyzed the samples using an AMS method. The AMS method allowed for increased sensitivity of the analyses (two to six times more sensitive than neutron activation). The increased sensitivity allowed for determining a background concentration of 0.0000009±0.0000002 pCi/L from a sample collected upgradient from the INTEC. The increased sensitivity resulted in detectable concentrations of ¹²⁹I downgradient of the INL that were used to calculate groundwater flow velocities of at least 6 ft/d. The maximum concentration detected in 1990–91 samples was 3.82±0.19 pCi/L, which was similar to the maximum concentration detected in 1986; however, mean concentrations from 18 wells sampled in 1986 and 1990-91 decreased from 1.30±0.26 to 0.81±0.19 pCi/L (Mann and Beasley, 1994b). This decrease was attributed largely to a decrease in disposal rates.

In 1992, Mann and Beasley (1994a) collected groundwater and surface water samples from 16 sites not likely to have been affected by wastewater disposal at the INTEC to determine background concentrations of ¹²⁹I. Concentrations of ¹²⁹I in water from nine wells, four springs, and three streams on (or tributaries to) the ESRP ranged from 0.0000001±0.0000001 to 0.0000081±0.0000006 pCi/L (average of 0.0000033±0.0000021 pCi/L). At the 99-percent confidence level, background concentrations of ¹²⁹I for the 16 sites were estimated to be less than or equal to 0.0000082 pCi/L. Cecil and others (2003) reevaluated the background concentrations by analyzing results of 52 samples (includes the samples collected by Mann and Beasley [1994a]) collected from groundwater and surface water during 1992–94 from various locations in the ESRP in southeastern Idaho. Cecil and others (2003) determined that surface water samples generally contained larger ¹²⁹I concentrations than groundwater samples because of anthropogenic fallout and evapotranspiration. They determined background concentrations using a subset of 30 groundwater samples from wells analyzed to be 0.0000054 pCi/L, and the 95-percent nonparametric confidence interval was 0.0000052–0.00001 pCi/L.

Hall (2006) collected samples from 13 wells downgradient of the INL during 1997 and 1998. Using AMS methods, Hall (2006) determined that concentrations in at least four of the wells (USGS 11, 14, 124, and 125; fig. 2) were greater than estimated background concentrations, and he postulated that 1958 peak ¹²⁹I in the ESRP aquifer had already passed these wells. Concentrations of these four wells ranged from 0.0000061±0.00000018 to 0.00074±0.00003 pCi/L, but they were less than concentrations measured in 1991 and 1993.

INL contractors routinely collect ¹²⁹I from monitoring wells throughout the INL for their Waste Area Group monitoring programs at the INL. In 2002, samples were collected from four wells south of the INTEC (ICPP 1795, 1796, 1797, and 1798; figs. 2 and 3) from three zones in the aquifer to determine concentrations above and below the H-I interbed (U.S. Department of Energy, 2004). Concentrations of ¹²⁹I in well ICPP-1795 increased from 0.34±0.04 pCi/L at 560 ft below land surface to 0.43±0.07 pCi/L at 620 ft below land surface. The three wells (ICPP 1796, 1797, 1798) farther to the south showed a decrease in ¹²⁹I concentration with depth, with concentrations in the upper zone ranging from 0.58±0.1 to 0.88±0.08 pCi/L and concentrations in the lower zone ranging from not detected to 0.33±0.05 pCi/L. The U.S. Department of Energy (2004, fig. 5-5) also presented results from analyses of 49 wells sampled in 2001; concentrations ranged from less than the method detection level of approximately 0.1 to 1.06 pCi/L. Analyses were completed using gamma spectroscopy methods. Concentrations for 20 wells sampled in 2003 were all less than the MCL of 1 pCi/L (U.S. Department of Energy, 2004, fig. 6-1).

Forbes and others (2007) presented results for 25 wells sampled in 2006 near the INTEC; concentrations ranged from less than the reporting level to 0.65 ± 0.097 pCi/L in USGS 67. Analyses of data collected from 2004–06 at wells around the INTEC indicated no discernible change in the concentrations when the uncertainty of the data was taken into account.

U.S. Department of Energy (2007) presented results for ¹²⁹I data collected in 2005 and 2006 from 24 wells downgradient of the INTEC and the ATRC (including several wells south of the INL), along with results from five zones each from two wells equipped with WestbayTM packer sampling systems (Middle 2050A and 2051). Samples were analyzed using the AMS method at the PRIME laboratory in Indiana, and some comparison was made to the 2003 USGS data presented in Bartholomay (2009). Results were used to speculate on the source of ¹²⁹I in wells around the Radioactive Waste Management Complex (RWMC). Concentrations in most southern wells were higher than background concentrations.

From 2005 to 2007, the Idaho Department of Environmental Quality (IDEQ) INL Oversight Program collected samples for 18 locations downgradient from the INL (Flint Hall, IDEQ, written commun., August 19, 2013). Samples were analyzed by the PRIME laboratory and concentrations ranged from 0.000000072±0.000000008 to 0.00000216±0.000000092. All the sample concentrations were within expected background concentrations.

U.S. Department of Energy (2008) presented results for ¹²⁹I data collected in 2007 from six zones in one WestbayTM equipped well (USGS 132). Samples were analyzed using the AMS method at the PRIME laboratory in Indiana, and the results from the six zones ranged from 0.0004±0.000013 to 0.002±0.00009 pCi/L. Results for all six zones were more than two orders of magnitude less than the MCL.

Bartholomay (2009) presented results for ¹²⁹I data collected from 36 wells in 2003 and 2007 along with concentrations from 31 zones sampled from six wells equipped with multilevel monitoring systems. Samples were analyzed using the AMS method at the PRIME laboratory in Indiana and concentrations ranged from 0.0000066±0.0000002 to 1.16 ± 0.04 pCi/L. The report highlighted concentration increases and decreases through time at various areas of the INL; decreases were attributed to discontinued disposal and dilution and dispersion in the aquifer. Increases were attributed to variable discharge rates of wastewater that eventually moved to well locations as a mass of water from a particular disposal period, and from the possible movement of remnant concentrations in perched aquifers around INTEC being moved to the ESRP aquifer.

U.S. Department of Energy (2012) presented results for ¹²⁹I data collected in 2011 from wells in and around INTEC. Concentrations were less than the MCL at all the monitoring wells and most of the wells had concentrations less than the laboratory detection levels. The highest concentration (0.537 pCi/L) was detected in well USGS 67. Trend plots indicated that ¹²⁹I concentrations declined significantly from concentrations detected during the 1980s and 1990s (U.S. Department of Energy, 2012, p. 27).

Methods and Quality Control

Sample Collection Methods

Sample collection by the USGS at the INL generally followed guidelines established by the USGS and documented in the USGS National Field Manual (U.S. Geological Survey, variously dated), and in Knobel and others (2008). Water was collected from wells with dedicated submersible pumps and from 11 wells equipped with dedicated WestbayTM packer sampling systems. The sample collected from the Kimama 460 well was collected with a thief sampler at 460 ft below land surface. The WestbayTM packer sampling systems allow for isolation of particular zones within the upper 30-650 ft of the aquifer, for sample collection. The other monitoring wells sampled for this study consist of open boreholes with variable completion depths in the upper 30-250 ft of the aquifer. The water sampled from these open boreholes often is a mixture of old, regional groundwater with young water that recharged at or near the INL (Busenberg and others, 2001).

Water from wells equipped with dedicated pumps was monitored during sampling for temperature, pH, and specific conductance using methods described by Wood (1981); Claassen (1982); and U.S Geological Survey (variously dated). Water samples were collected after field measurements stabilized and after at least 1 volume of water was purged from each well. Samples collected prior to October 2003 for previous studies were collected after 3 volumes of water were purged, and Bartholomay (1993) and Knobel (2006) determined that the difference between purging 1 and 3 wellbore volumes at selected INL wells had no discernible effect on statistical comparability of select water-quality data; however, ¹²⁹I data was not examined.

For wells with dedicated WestbayTM packer sampling systems, pre-cleaned stainless-steel thief sampling bottles were lowered to the zone to be sampled, connected to the sampling port, and filled with formation water. The filled stainless-steel bottles then were raised to the surface and emptied into a pre-cleaned container; the water was then processed to fill 1-L glass amber bottles. Field measurements also were taken from the pre-cleaned container.

Field processing of all samples consisted of filtering the water through a disposable 0.45-micrometer filter cartridge that had been pre-rinsed with at least 1 L of deionized water or 1 L of sample water. Filtration was necessary to remove particulate matter that could affect the laboratory preparation of the silver iodide targets used in the AMS measurements of ¹²⁹I (Cecil and others, 2003). In addition, a potassium hydroxide and sulfurous acid stabilizer solution was added to each sample so they could be stored until shipment and chemical preparation at the laboratory.

Analytical Methods

Iodine-129 concentrations in the groundwater samples were determined using AMS methods described by Sharma and others (1997) and quality-control requirements described by Mark Caffee, Purdue University (written commun., accessed September 10, 2013, at http://www.physics.purdue.edu/primelab/AMSQAQC/qaupdate.php). The AMS used to analyze the samples is located at the PRIME laboratory, Purdue University, West Lafayette, Indiana. Water samples go through a chemical process to produce a silver iodide target material. The silver iodide is prepared after the addition of an iodine carrier, and the target is placed in a holder for AMS analyses. Analyses of the target produce a ratio of ¹²⁹I to stable iodine-127 (¹²⁷I) for that sample.

Guidelines for Interpretation of Analytical Results

Concentrations of ¹²⁹I are reported with an estimated sample standard deviation, *s*, which is obtained by propagating sources of analytical uncertainty in measurements. The guidelines for interpreting analytical results are based on an extension of a method proposed by Currie (1984) and are given in Mann and Beasley (1994b). In this report, ¹²⁹I concentrations less than 3*s* are considered less than a "reporting level." The reporting level should not be confused with the analytical method detection limit, which is based on laboratory procedures.

Laboratory results from previous analyses (1990–91 and 2003) were reported by the laboratory as the ratio of ¹²⁹I to ¹²⁷I and converted to concentrations in picocuries per liter (pCi/L) by the authors for the 1990–91 data (Mann and Beasley, 1994b) and by using an equation described by Rao (1997) for the 2003 data:

$$S_{T} = \frac{[R_{m}(C_{c}C_{v} + S_{c}S_{v}) - C_{c}C_{v}C_{r}]N}{fS_{v}}$$
(1)

where

- S_T is the ¹²⁹I concentration in atoms per liter of water,
- R_m is the ratio of ¹²⁹I to ¹²⁷I measured by AMS,
- C_c is the concentration of iodine in grams per liter (g/L) of carrier solution,
- C_v is the volume of carrier solution added in L,
- C_r^{129} I to ¹²⁷ I ratio for carrier solution,
- S_c is the concentration of iodine in g/L for sample,
- S_{v} is the volume of the sample in L,
- *N* is Avogadro's number $(6.023 \times 10^{23} \text{ atoms per mole})$, and
- *f* is the formula weight for iodine (126.9 grams per mole).

The mass of the carrier given by the PRIME laboratory is the product of carrier volume (C_{ν}) and carrier concentration (C_c) in milligrams. The concentration of iodine in most samples was less than 0.002 mg/L, which was used for the concentration for all samples except those with estimated or reported concentrations.

For 2010–12, the laboratory reported results in ratios of ¹²⁹I to ¹²⁷I and in pCi/L based on the equation:

$$\frac{\text{pCi}}{\text{L}}(\text{sample}) = \frac{(R_s - R_b) \times (W_s + W_c) \times 1.80 \times 10^{-7}}{Wt} \left(\frac{\text{pCi}}{\text{L}}\right) \quad (2)$$

where

 R_s is the measured sample ratio of ¹²⁹I atoms to ¹²⁷I atoms,

 R_b is the ratio of the chemistry blank,

 W_s is the weight of ¹²⁷I in sample (in mg),

 W_c is the weight of ¹²⁷I added as carrier (in mg), and

Wt is weight of sample (aliquot).

The laboratory calculations do not take into account the concentration of iodine in the sample because the concentration typically is negligible; the concentrations were estimated in the equation (Rao, 1997) used for 2003 data. Differences between the reported concentrations are similar between both equations when sample ratios are larger than carrier blank ratios; however, when ¹²⁹I/¹²⁷I ratios of the samples are less than the ratio for the carrier blank sample, negative results occur for calculations based on the Rao (1997) equation. The PRIME lab did not subtract the carrier blank ratios for the samples that had carrier blank ratios larger than sample ratios (Susan Ma, PRIME lab, written commun., August 20, 2013).

Quality Control

Bias and variability were assessed with blank and replicate quality control (QC) samples submitted for AMS analyses with the standard samples. One source solution blank (QAW-34), one equipment blank (QAW-35), and six QC replicate samples were collected. The blanks consisted of inorganic-free water, obtained from the USGS National Water Quality Laboratory, that was passed through the same sample collection and processing equipment used for sample collection. Results for both blanks were less than background concentrations (table 1) indicating no bias of the ¹²⁹I concentrations from the field processing of the samples.

Statistical equivalency of radiochemical-constituent concentrations in sample replicate pairs was determined following a method defined by Volk (1969) and described in more detail by Williams (1996). In this method, statistical equivalence is determined within a specified confidence level. A value for the standard deviate, *Z*, is calculated, and then the level of significance of the result is evaluated (evaluation of the level of significance assumes that the sample population is distributed normally). For this report, concentrations of individual constituents in sample pairs (constituent pairs) were considered equivalent when the results were within 2 standard deviations of each other. At this confidence level (95-percent), the level of significance, determined from a standard normal probability curve, was 0.05 for a two-tailed test, and it corresponded to a *Z*-value of 1.96.

The equation used to determine *Z* was adapted from Volk (1969):

$$Z = \frac{|x - y|}{\sqrt{(s_x)^2 + (s_y)^2}}$$
(3)

where

- *x* is the concentration of a constituent in the routine sample,
- *y* is the concentration of the same constituent in the sequential replicate sample,
- s_x is the standard deviation of x, and,
- s_y is the standard deviation of y.

When the population is not distributed normally, which often is the case with radiochemical results (L. DeWayne Cecil, U.S. Geological Survey, written commun., January 4, 2009), or an approximation of the standard deviation is used, a *Z*-value less than 1.96 must be considered as a guide when testing for equivalence. Constituent concentrations in sample pairs were considered statistically equivalent when the calculated *Z*-value was less than or equal to 1.96.

The use of equation 3 therefore is considered a guide in determining if the results of ¹²⁹I analyses of a replicate pair of samples were equivalent. The results and reported standard deviations for the analyses of ¹²⁹I replicate pairs and the *Z*-values are listed in <u>tables 1</u> and <u>2</u>. Results for four of the six replicate pairs had *Z*-values less than 1.96 and can be considered statistically equivalent.

The remaining two samples had results not considered statistically equivalent using equation 3. Both pairs of the remaining two samples had low concentrations and their uncertainties were an order of magnitude smaller than the concentrations which caused them to have Z-values greater than 1.96. Results of blank and replicate samples generally indicated that the sample collection and laboratory procedures used were appropriate for the type of samples and range of concentration observed. However, given the extreme sensitivity of the AMS analysis method and the range of concentrations detected, as the ¹²⁹I concentration approached background levels and the concentration of ¹²⁹I in the blank samples. Z-values to determine statistical significance of replicate pairs tended to increase. This potentially raises some minor concern regarding statistically significant reproducibility at low concentration levels near or within background levels.

 Table 1.
 Iodine-129 concentrations in groundwater, Idaho National Laboratory and vicinity, Idaho, 2010–2012.

[See figure 2 for well locations. **Concentration:** Concentrations and analytical uncertainties are in picocuries per liter; uncertainties are 1*s*. **Z-value** is the statistical test used to compare the replicate values. **Abbreviations:** NA, not applicable; QA, quality assurance; QAI, quality assurance iodine; QAW, quality assurance blanks]

Sample	Site	Date	Concentration	7-value
identifier	identifier	sampled	ooncentration	2 Value
CFA-1	433204112562001	04-18-12	0.37±0.017	0.08
QAI-3	433204112562001	04-18-12	0.368±0.02	NA
CFA-2	433144112563501	10-17-11	0.0798 ± 0.0024	NA
CFA LF 2-10	433216112563301	04-18-12	0.177±0.017	NA
Crossroads	432128113092701	04-23-12	0.0000066 ± 0.0000009	NA
Kimama 460	425023113474301	10-07-10	0.0000031 ± 0.0000006	NA
Kimama WS	425023113474401	10-06-10	0.0000013 ± 0.0000005	NA
NRF-6	433910112550101	11-07-11	0.000113 ± 0.000004	NA
NRF 8	433843112550901	11-08-11	0.000011 ± 0.0000008	NA
NRF 9	433840112550201	11-08-11	0.000025 ± 0.0000016	NA
NRF 10	433841112545201	11-09-11	0.0000128 ± 0.0000011	3.82
QA-2	433841112545201	11-09-11	0.0000193 ± 0.0000013	NA
NRF 11	433847112544201	11-09-11	0.0000143 ± 0.000001	NA
Rifle Range	433243112591101	10-17-11	0.00231 ± 0.00012	NA
RWMC Prod.	433002113021701	10-13-11	0.0162 ± 0.0005	NA
USGS 9	432740113044501	10-18-11	0.0000107 ± 0.0000007	NA
USGS 11	432336113064201	04-16-12	0.000046 ± 0.0000025	NA
USGS 14	432019112563201	10-19-11	0.0000177 ± 0.000001	NA
USGS 20	433253112545901	04-03-12	0.037±0.0021	NA
USGS 34	433334112565501	04-04-22	0.0056 ± 0.0004	NA
USGS 35	433339112565801	10-05-11	0.0007 ± 0.00005	NA
USGS 36	433330112565201	04-04-12	0.035 ± 0.005	NA
USGS 37	433326112564801	10-05-11	0.31±0.016	NA
USGS 38	433322112564301	04-04-12	0.202±0.018	NA
USGS 41	433409112561301	10-26-11	0.135 ± 0.007	NA
USGS 42	433404112561301	04-05-12	0.214±0.013	NA
USGS 43	433415112561501	10-06-11	0.101 ± 0.004	NA
USGS 44	433409112562101	04-03-12	0.112±0.005	NA
USGS 45	433402112561801	10-06-11	0.359±0.012	NA
USGS 47	433407112560301	10-26-11	0.349±0.019	NA
USGS 48	433401112560301	04-05-12	0.215 ± 0.018	NA
USGS 51	433350112560601	04-02-12	0.151 ± 0.01	NA
USGS 52	433414112554201	10-12-11	0.165 ± 0.008	NA
USGS 57	433344112562601	10-11-11	0.333±0.021	NA
USGS 59	433354112554701	04-04-12	0.2±0.01	NA
USGS 67	433344112554101	10-06-11	1.02 ± 0.04	0.71
QA-1	433344112554101	10-06-11	1.06 ± 0.04	NA
USGS 77	433315112560301	10-06-11	0.153 ± 0.007	NA
USGS 82	433401112551001	04-02-12	0.007 ± 0.0005	NA
USGS 85	433246112571201	04-03-12	0.113±0.008	NA
USGS 104	432856112560801	10-19-11	0.027 ± 0.007	NA
USGS 106	432959112593101	10-19-11	0.0288 ± 0.0012	NA
USGS 111	433331112560501	04-02-12	0.166 ± 0.009	NA
USGS 112	433314112563001	10-11-11	0.43±0.03	NA
USGS 113	433314112561801	04-02-12	0.477±0.026	0.56
QAI-2	433314112561801	04-02-12	0.499±0.029	NA
USGS 114	433318112555001	10-12-11	0.141 ± 0.004	NA
USGS 115	433320112554101	10-11-11	0.0124 ± 0.0004	NA
USGS 116	433331112553201	06-06-12	0.242 ± 0.011	NA

 Table 1.
 Iodine-129 concentrations in groundwater, Idaho National Laboratory and vicinity, Idaho, 2010–2012.—Continued

[See figure 2 for well locations. **Concentration:** Concentrations and analytical uncertainties are in picocuries per liter; uncertainties are 1*s*. **Z-value** is the statistical test used to compare the replicate values. **Abbreviations:** NA, not applicable; QA, quality assurance; QAI, quality assurance iodine; QAW, quality assurance blanks]

Sample identifier	Site identifier	Date Concentration		Z-value
USGS 123	433352112561401	10-11-11	0.0342±0.002	NA
USGS 124	432307112583101	04-16-12	0.00186 ± 0.0001	NA
USGS 125	432602112052801	10-18-11	0.000293 ± 0.00001	NA
USGS 127	433058112572201	06-06-12	0.00086 ± 0.00005	NA
USGS 128	433225112565601	10-05-11	0.0927±0.0023	NA
USGS 136	433447112581501	10-25-11	0.00292 ± 0.00008	NA
QAW-34	433000113000001	10-24-12	0.00000044 ± 0.0000029	NA
QAW-35	433000113000001	10-24-12	$0.00000074 \pm 0.00000028$	NA

Table 2. Iodine-129 concentrations at multiple aquifer depths at the Idaho National Laboratory, Idaho, 2012.

[QAW-36 is a replicate of USGS 131A Zone 8, QAW-33 is a replicate of USGS 137A Zone 3, QAW-34 is a source solution blank, and QAW-35 is an equipment blank. **Port depth** in feet below land surface. **Z-value** is the statistical test used to compare the replicate samples. **Abbreviations:** pCi/L, picocuries per liter; NA, not applicable; QAW,quality assurrance westbay; NA, not applicable]

Sample identifier	Site identifier	Port depth	Date sampled	lodine-129 (pCi/L)	Z-value
Middle 2050A Zone 15	433409112570515	517	06-19-12	0.000006±0.000004	NA
Middle 2051 Zone 3	433217113004903	1,091	06-20-12	0.00507 ± 0.00021	NA
Middle 2051 Zone 9	433217113004909	749	06-20-12	0.00057 ± 0.00008	NA
USGS 103 Zone 1	432714112560702	1,258	06-25-12	0.0117±0.0007	NA
USGS 103 Zone 3	432714112560704	1,210	06-25-12	0.0112±0.0007	NA
USGS 103 Zone 6	432714112560708	1,087	06-25-12	0.0111±0.0007	NA
USGS 103 Zone 9	432714112560712	993	06-25-12	0.0077 ± 0.0006	NA
USGS 105 Zone 5	432703113001807	1,070	06-28-12	0.00453 ± 0.00012	NA
USGS 105 Zone 8	432703113001811	949	06-28-12	0.00471 ± 0.00012	NA
USGS 105 Zone 11	432703113001815	849	06-28-12	0.00216 ± 0.0001	NA
USGS 108 Zone 1	432659112582602	1,172	06-26-12	0.00544 ± 0.00025	NA
USGS 108 Zone 4	432659112582606	1,029	06-26-12	0.00638 ± 0.00028	NA
USGS 108 Zone 7	432659112582610	888	06-26-12	0.00513±0.00013	NA
USGS 131A Zone 3	433036112581803	1,137	10-24-12	0.0099 ± 0.0005	NA
USGS 131A Zone 5	433036112581806	981	10-28-12	0.0095 ± 0.0006	NA
USGS 131A Zone 8	433036112581810	812	10-28-12	0.082±0.003	0.34
QAW-36	433036112581810	812	10-28-12	0.084 ± 0.005	NA
USGS 131A Zone 12	433036112581815	616	10-28-12	0.0151 ± 0.0008	NA
USGS 132 Zone 14	432906113025018	765	06-19-12	0.00105 ± 0.00004	NA
USGS 133 Zone 10	433605112554312	469	06-21-12	0.000029 ± 0.000003	NA
USGS 134 Zone 9	433611112595811	707	06-18-12	0.0005 ± 0.0004	NA
USGS 135 Zone 7	432753113093609	837	06-21-12	0.000029 ± 0.000003	NA
USGS 137A Zone 1	432701113025801	876	10-23-12	0.000344 ± 0.000024	NA
USGS 137A Zone 3	432701113025803	841	10-23-12	0.000359 ± 0.000012	2.49
QAW-33	432701113025803	841	10-23-12	0.000315 ± 0.000013	NA
USGS 137A Zone 4	432701113025805	747	10-23-12	0.00102 ± 0.00004	NA
USGS 137A Zone 5	432701113025807	662	10-24-12	0.00139 ± 0.00005	NA
QAW-34	433000113000001	NA	10-24-12	$0.00000044 \pm 0.00000029$	NA
QAW-35	433000113000001	NA	10-24-12	0.00000074±0.00000028	NA

Concentrations of Iodine-129 in the Eastern Snake River Plain Aquifer

In October 2010, water samples were collected from 2 wells located near Kimama, Idaho (fig. 1) to determine whether or not water from the INL could be affecting water quality in that area of the ESRP. Both wells (Kimama 460; 0.0000031 ± 0.0000006 pCi/L and Kimama WS; 0.0000013 ± 0.0000005 pCi/L; table 1) had lower ¹²⁹I concentrations than what is considered to be the background concentration in the ESRP aquifer (Cecil and others, 2003), so the INL does not appear to be affecting the water quality at that location.

From October 2011 through April 2012, water samples were collected from 49 ESRP aquifer wells at or south of the INL. The areal distribution of concentrations greater than 0.1 pCi/L is shown in figure 4. Concentrations of ¹²⁹I in the aquifer ranged from 0.0000066±0.0000009 to 1.02±0.004 pCi/L (table 1, fig. 4). Water from one well south of the INL (Crossroads) had a concentration (0.0000066 pCi/L) that was within the uncertainty of the estimated background concentration of 0.0000054 pCi/L for the ESRP aquifer as calculated by Cecil and others (2003) and was less than the background concentration of 0.0000082 pCi/L as calculated by Mann and Beasley (1994a).

During 2011, samples for ¹²⁹I analyses were collected from five wells (NRF 6, 8, 9, 10, and 11; fig. 1) around the NRF, that had never been previously sampled. Concentrations ranged from 0.000011 \pm 0.0000008 pCi/L in NRF 8 to 0.000113 ±0.000004 pCi/L in NRF 6 and are all slightly higher than what would be considered background levels in ESRP aquifer. Disposal records from NRF could not be found that indicated disposal of ¹²⁹I in wastewater at the facility; however, it is possible there was some unknown amount of ¹²⁹I in wastewater discharged in the past. One other possible source, other than unknown wastewater disposal at NRF, could be air pollutant source emissions from INTEC. Iodine-129 has been present in air emissions throughout INTEC's history (Taylor, 1994) and the predominant windflow direction at the INL during the day is from the southwest to northeast (Abbott and others, 2004).

Concentrations of ¹²⁹I in the ESRP aquifer generally decreased from 1990–91 to 2011–12 (<u>table 3</u>). The average concentration of 15 wells sampled during four different sample periods decreased from 1.15 pCi/L in 1990–91 to 0.27 in 2003; 0.252 in 2007; and 0.173 pCi/L in 2011–12. Concentrations of ¹²⁹I in wells within a 3-mi radius of the INTEC decreased as much as one order of magnitude in concentration (<u>table 3</u>). These decreases are attributed to discontinuation of disposal of ¹²⁹I in wastewater after 1988 and to dilution and dispersion in the ESRP aquifer.

Although the wells near the INTEC showed decreases of ¹²⁹I, wells at Central Facilities Area (CFA) (CFA LF 2-10 and CFA-1); south and southeast of the CFA (USGS 127, USGS 104, and USGS 106); near the site boundary (USGS 103 and 108); and south of the INL (USGS 11 and USGS 125) showed slight increases in concentrations between 2003 or 2007 and 2011-12 (table 3, fig. 5). However, some of the wells south of INTEC also showed slight decreases (CFA-2 and USGS 105, 124 and 14). These slight increases or decreases may be related to variable discharge rates of wastewater and wastewater concentration that moved to these well locations as a mass of water from a particular disposal period. For example, the highest concentrations of ¹²⁹I probably were in wastewater discharged in 1957 (U.S. Department of Energy 2004, appendix D) and 1978 (Mann and Beasley, 1994b, fig. 2; U.S. Department of Energy, 2004, appendix D). Therefore, when water reaches a well from discharge in 1957 and 1978, the concentration of the sample most likely would be greater than concentrations in samples of water discharged during other periods. Data from wells USGS 11 and USGS 14 from Hall (2006) somewhat support this idea because ¹²⁹I concentrations for these two wells were less in 1998 than during the 1990-91 and 2003 sample periods. Cecil and others (2000) indicated that 1958 peak disposal of chlorine-36 probably reached USGS 11 in 1984 and reached USGS 14 in 1987. Beasley and others (1998) examined the relative mobility of several isotopes disposed at the INTEC, and they concluded that chlorine-36 behaves conservatively in the basalt; however ¹²⁹I is attenuated by sorption with other ionic species, and probably moves more slowly in the system. Sample results for 1990-91 from some southern wells, therefore, could represent the end of high concentrations related to the large discharge event in 1957; 2003 and 2007 data could represent the beginning or end of high concentrations related to the 1978 discharge event or possibly some other discharge period depending on the age and travel time of the young fraction of water; and 2011-12 data may be representative of discharge for some time after 1978. Busenberg and others (2001, table 8 and fig. 25) calculated the age of the young fraction of groundwater and relative flow rates for several of the wells, at and south of the INL; and the age and flow velocity to USGS 11 and USGS 125 were younger and faster than the age and flow velocity to USGS 124 and USGS 14, which could account for the observed increase in concentrations for USGS 11 and USGS 125 and decrease for USGS 124 and USGS 14. Accurately predicting the first arrival of peak concentrations with the limited sample periods is difficult because of the uncertainty of ¹²⁹I concentrations in wastewater discharged prior to 1976 and the complexity of flow movement in the basaltic aquifer system.



Universal Transverse Mercator projection, Zone 12 Datum is North American Datum of 1927

Figure 4. Areal distribution of iodine-129 in the eastern Snake River Plain aquifer at the Idaho National Laboratory, Idaho, 2011–12.

Table 3. Concentrations of iodine-129 in water from selected wells, Idaho National Laboratory and vicinity, Idaho, 1990–2012.

[See figures 2 and 3 for well locations. Concentrations and analytical uncertainties are in picocuries per liter; uncertainties are 1*s*. Average concentration calculated from the 15 wells with concentrations from all four sample periods (USGS 123 excluded because well was modified in 2004). **Abbreviation:** NS, not sampled]

Sample identifier	1990–91	2003	2007	2011–12
CFA-1	0.24±0.05	NS	0.318±0.015	0.37±0.017
CFA-2	0.10±0.03	NS	0.131±0.006	0.0798 ± 0.0024
CFA LF 2-10	NS	0.079±0.003	NS	0.177±0.017
Rifle Range	NS	NS	0.001+0.004	0.00231 ± 0.00012
RWMC Prod.	NS	NS	0.0188+0.0005	0.0162+0.0005
USGS 9	NS	0.0000088±0.00000035	NS	0.0000107±0.0000007
USGS 11	0.00001 ± 0.000001	0.000018+0.0000005	NS	0.000046+0.0000025
USGS 14	0.00003 ± 0.000002	0.00004 ± 0.000002	NS	0.0000177 ± 0.000001
USGS 20	0.033±0.002	0.026±0.0011	0.0282 ± 0.0009	0.037±0.0021
USGS 34	0.39+0.02	NS	0.0098+0.001	0.0056+0.0004
USGS 35	0.57+0.03	0.0018+0.00002	NS	0.0007 ± 0.00005
USGS 36	1.19 ± 0.03	0.162 ± 0.008	NS	0.035±0.005
USGS 37	1.80 ± 0.08	0.452±0.025	0.395±0.017	0.31±0.016
USGS 38	2.00+0.07	0.556+0.019	0.281+0.008	0.202+0.018
USGS 41	0.58±0.03	NS	NS	0.135±0.007
USGS 42	3.82 ± 0.19	0.216±0.0064	0.325±0.01	0.214±0.013
USGS 43	0.16 ± 0.01	NS	NS	0.101±0.004
USGS 44	0.20±0.03	0.12±0.0035	NS	0.112±0.005
USGS 45	0.32 ± 0.01	NS	NS	0.359±0.012
USGS 47	0.83 ± 0.04	0.621±0.022	NS	0.349±0.019
USGS 48	0.22 ± 0.02	NS	0.35±0.013	0.215±0.018
USGS 51	0.28+0.01	0.164+0.0071	0.231+0.01	0.151+0.01
USGS 52	0.38±0.03	NS	0.284 ± 0.014	0.165 ± 0.008
USGS 57	1.38+0.07	0.64+0.023	0.521+0.023	0.333+0.021
USGS 59	0.57+0.02	NS	0.262+0.013	0.2+0.01
USGS 67	1.43+0.04	NS	1.16+0.04	1.02+0.04
USGS 77	1.37+0.06	0.586+0.0193	0.71+0.04	0.153+0.007
USGS 82	0.119+0.002	0.0112+0.0004	0.011+0.0004	0.007 ± 0.0005
USGS 85	1.64+0.08	0.283+0.009	0.173+0.006	0.113+0.008
USGS 104	0.0036+0.0001	0.0049+0.0002	0.005 ± 0.00023	0.027+0.007
USGS 106	0.025+0.001	0.034+0.001	0.0274+0.0013	0.0288+0.0012
USGS 111	0.86+0.09	0.138+0.007	0.192 ± 0.007	0.166+0.009
USGS 112	2.40+0.25	0.617+0.067	NS	0.43+0.03
USGS 113	3.25+0.14	0.72+0.051	0.75+0.04	0.477+0.026
USGS 114	0.28+0.01	0.153+0.0063	0.173+0.006	0.141+0.004
USGS 115	0.19+0.02	0.0172+0.0005	NS	0.0124+0.004
USGS 116	0.45+0.01	0.069+0.0026	0.144+0.005	0.242+0.011
USGS 123	1.00+0.05	0.678+0.034	0.048+0.004	0.0342 ± 0.002
USGS 124	NS	0.0023+0.0001	0.00225+0.00007	0.00186+0.0001
USGS 125	NS	0.00013 ± 0.0000008	NS	0.000293±0.00001
USGS 127	NS	0.001 ± 0.00008	0.00071+0.00003	0.00086 ± 0.00005
USGS 128	NS	0.263 ± 0.0085	0.162 ± 0.008	0.0927 ± 0.0023
Average concentration	1.15	0.27	0.252	0.173



Figure 5. Change in concentration of iodine-129 between samples collected during 2003 or 2007 and 2011-12.

Bartholomay (2009) compared ¹²⁹I concentrations from 2003 and 2007 between several wells around INTEC and noted areas where concentrations seemed to be decreasing and areas where concentrations appeared to be increasing. In the wells with concentrations that increased, Bartholomay (2009) concluded that the 2007 increase could be due to flow in the Big Lost River during 2005 and 2006 moving remnant perched water concentrations into the aquifer. A similar comparison was done between the previous sample collected and the 2011–12 data (fig. 6). Figure 6 shows that ¹²⁹I decreased in most of the wells, except USGS 45 and USGS 116, since the previous sampling event. The decreasing concentrations are consistent with discontinued disposal in the area. Some flow did occur at the INL in the Big Lost River at Lincoln Boulevard bridge (13132535, fig. 1) during 2009–12 (10,437 acre-ft), but the flow was much less than what occurred during water year 2006 alone (17,340 acre-ft) (access annual water data at http://maps.waterdata.usgs. gov/mapper/index.html [U.S. Geological Survey, 2013b]), so if recharge was the primary reason for the increases in 2007 the decrease in recharge (along with normal dilution and dispersion) during the 2007-12 timeframe could be the reason for the decrease in concentrations. Mirus and others (2011) indicated that streamflow provides local recharge to the shallow, intermediate, and deep perched zones within about 450 ft of the Big Lost River and indicated other perched water dynamics including local snowmelt and anthropogenic sources (such as leaky pipes and drainage ditches) contribute to recharge of shallow and intermediate perched zones throughout much of INTEC. It is not clear as to why the concentrations may have increased in USGS 45 and 116; however, the concentration from USGS 45 only had a 1990 value to compare with and USGS 116 had variable decreases and increases between the four sample periods (table 3).

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

During June and October 2012, water samples were collected from 25 zones from 11 wells equipped with multi-level WestbayTM packer sampling systems (Middle 2050A, Middle 2051, and USGS 103, 105, 108, 131A, 132, 133, 134, 135, and 137A; figs. 2 and 3). The MLMS isolate various zones of the aquifer so the relative vertical distribution of ¹²⁹I can be determined. Water from all but one of the sampled zones contained concentrations that were greater than the reporting level of 3*s*, and concentrations ranged from 0.000006 ± 0.000004 to 0.082 ± 0.003 pCi/L (table 2). All concentrations were greater than the estimated background concentration of 0.0000054 pCi/L for the ESRP aquifer as calculated by Cecil and others (2003), although the sample from Middle 2050A Zone 15 was within the uncertainty of the background concentration (table 2). The two new wells (USGS 131A and USGS 137A), which were completed in August 2012 and had samples collected from all four sample zones, showed variability of up to one order of magnitude in ¹²⁹I concentrations among various zones. The upper two zones in USGS 131A and USGS 137A had larger concentrations than the two deeper zones (table 2). Two other wells (USGS 105 and USGS 108), that were completed after the 2007 ¹²⁹I study, had three sample zones collected and all three zones from each well showed similar concentrations (table 2). One other well (USGS 135) also was completed after the 2007 study, and only one zone was sampled because previous water chemistry from the well indicated similar chemistry in all four of its sample zones (access data at http://waterdata.usgs.gov/nwis/inventory?agency code=USGS&site_no=432753113093602 [U.S. Geological Survey, 2013c]). Concentrations were well below the MCL in all zones from all the wells.

Bartholomay (2009) first described ¹²⁹I concentrations in six of the wells with MLMS installed through 2007. Table 4 lists the wells and zones that were sampled in 2007 and 2012 to see whether or not concentrations were increasing or decreasing in the respective zones, figures 5 and 6 denote the change spatially. Three of the MLMS wells sampled in 2012 were either monitoring wells in 2007 (USGS 105 and USGS 108) or had a monitoring well in close proximity (USGS 109 for comparison with USGS 137A) that were used to compare the mixed water chemistry of the approximate upper 150 ft of the aquifer with the upper MLMS zone of water. The comparison between similar zones for ¹²⁹I concentrations in six (Middle 2050A, Middle 2051, and USGS 105, 132, 133, and 134) of the nine wells indicated concentrations decreased in 2012. The wells that showed increases were located along the southern boundary of the INL (USGS 137A, USGS 108, and USGS 103; fig. 5). The decreasing concentrations can be attributed to dilution and dispersion in the aquifer system. All three zones in USGS 103 that were sampled in 2012 showed an increase of ¹²⁹I concentrations by an order of magnitude from the 2007 study. These zones historically have had variable tritium concentrations greater than the reporting level, and are influenced more by wastewater disposal than the other zones in the well, so the increases are probably related to variable discharge of wastewater in the past at INTEC. The other increases in USGS 108 and USGS 137A also could be due to variable wastewater discharge or to changes in the portion of the aquifer represented by the samples used for the comparison.



Base from U.S. Geological Survey digital data, 1:24,000 and 1:100,000 Universal Transverse Mercator projection, Zone 12 Datum is North American Datum of 1927

Figure 6. Change in concentration of iodine-129 between samples collected between 1990–2007 and 2011–12.

The variability with some ¹²⁹I concentrations with depth in the new wells installed in 2012 (USGS 131A and USGS 137A) is consistent with the variability of some of the other water chemistry in the wells (<u>table 5</u>). For example, samples from USGS 131A, Zone 8, at 812 ft below land surface had the highest ¹²⁹I concentration, and had chloride and tritium concentrations greater than the other zones (<u>table 5</u>). Concentrations of these constituents are related to wastewater disposal at INTEC. Concentrations of ¹²⁹I are higher in the upper two zones of USGS 137A but the other water chemistry appears to be consistent in all four zones. Chloride concentrations in samples from the upper zone (zone 5 at 662 ft below land surface) was slightly higher (14.3 mg/L) than the other three zones (11.5–12.4 mg/L). The higher concentrations of ¹²⁹I and chloride probably are due to more influence of wastewater disposal at INTEC in that part of the aquifer.

Table 4.Iodine-129 concentrations at multiple aquifer depths at the IdahoNational Laboratory, Idaho, 2007 and 2012.

[2007 samples (Bartholomay, 2009) for USGS 105, 108, and 109 from about upper 150 feet of open boreholes were used to compare with upper zones of multilevel wells USGS 105 zone 11, 108 zone 7, and 137A zone 5, respectively. Concentrations in picocuries per liter. **Port depth** in feet below land surface. Port depth differs from Bartholomay (2009) because of remeasurement. **Abbreviations:** NA, not applicable]

Sample identifier	Port depth	2007	2012
Middle 2050A Zone 15	517	0.00019±0.00005	0.000006±0.000004
Middle 2051 Zone 3	1,091	0.0066 ± 0.00022	0.00507 ± 0.00021
Middle 2051 Zone 9	749	0.00079 ± 0.00005	0.00057 ± 0.00008
USGS 103 Zone 1	1,258	0.0012 ± 0.00004	0.0117 ± 0.0007
USGS 103 Zone 3	1,210	0.00046 ± 0.00003	0.0112 ± 0.0007
USGS 103 Zone 6	1,087	0.0017 ± 0.0014	0.0111±0.0007
USGS 105	NA	0.00227 ± 0.00009	NA
USGS 105 Zone 11	849	NA	0.00216 ± 0.0001
USGS 108	NA	0.00102 ± 0.00004	NA
USGS 108 Zone 7	888	NA	0.00513 ± 0.00013
USGS 132 Zone 14	765	0.00111 ± 0.00007	0.00105 ± 0.00004
USGS 133 Zone 10	469	0.0011 ± 0.00024	0.000029 ± 0.000003
USGS 134 Zone 9	707	0.0167 ± 0.0007	0.0005 ± 0.0004
USGS 109	NA	0.000677 ± 0.00002	NA
USGS 137A Zone 5	662	NA	0.00139 ± 0.00005
USGS 137A Zone 4	747	NA	0.00102 ± 0.00004

Table 5. Concentrations of selected chemicals at multiple aquifer depths from two wells at the Idaho National Laboratory, Idaho, 2012.

[**Port depth** in feet below land surface. **Abbreviations**: pCi/L, picocuries per liter, uncertainty is 1*s*; Ca, calcium; Na, sodium; HCO³, bicarbonate; Cl, chloride; ²H, deuterium; ¹⁸O, oxygen-18; permil, parts per thousand, relative to standard mean ocean water; mg/L, milligrams per liter]

Sample identifier	Site identifier	Port depth	Date sampled	lodine-129 (pCi/L)	Ca (mg/L)	Na (mg/L)	HCO ³ (mg/L)	Cl (mg/L)	² H (permil)	¹⁸ 0 (permil)	Tritium (pCi/L)
USGS 131A Zone 3	433036112581803	1,137	10-24-12	0.0099±0.0005	48.4	7.87	202	14	-137	-18.04	140±60
USGS 131A Zone 5	433036112581806	981	10-28-12	0.0095±0.0006	47.9	8.33	200	13.5	-137	-18.00	150±60
USGS 131A Zone 8	433036112581810	812	10-28-12	0.082±0.003	53.7	10.3	180	26	-136	-17.84	$1,590 \pm 90$
USGS 131A Zone 12	433036112581815	616	10-28-12	0.0151±0.0008	45.9	8.3	171	16.1	-138	-17.96	940±70
USGS 137A Zone 1	432701113025801	876	10-23-12	0.000344 ± 0.000024	41	10.6	177	11.9	-136	-17.82	140±60
USGS 137A Zone 3	432701113025803	841	10-23-12	0.000359 ± 0.000012	41.2	10.6	173	11.5	-138	-17.83	100±60
USGS 137A Zone 4	432701113025805	747	10-23-12	0.00102 ± 0.00004	40.9	11.1	173	12.4	-138	-17.86	130±60
USGS 137A Zone 5	432701113025807	662	10-24-12	0.00139 ± 0.00005	38.6	11.4	174	14.3	-137	-17.86	100 ± 50

Summary

From 1953 to 1988, wastewater containing approximately 0.94 curies of iodine-129 (¹²⁹I) was generated at the Idaho National Laboratory (INL) in southeastern Idaho. Almost all this wastewater was discharged at or near the Idaho Nuclear Technology and Engineering Center (INTEC) on the INL site. Most of the wastewater was discharged directly into the eastern Snake River Plain (ESRP) aquifer through a deep disposal well until 1984; however, some wastewater also was discharged into unlined infiltration ponds or leaked from distribution systems below the INTEC.

The U.S. Geological Survey (USGS) conducted monitoring programs for ¹²⁹I in the ESRP aquifer at the INL in 1977, 1981, 1986, 1990–91, 2003, and 2007 prior to sample collection in 2010–12. Some additional sampling was done in the 1990s to determine an estimated background concentration of the ESRP aquifer water of 0.0000054 picocuries per liter (pCi/L). Current ¹²⁹I concentrations in the ESRP aquifer at the INL are compared to the background concentration and to the U.S. Environmental Protection Agency (USEPA) maximum contaminant level (MCL) of 1 pCi/L.

In 2010–12, the USGS, in cooperation with the U.S. Department of Energy, collected samples for ¹²⁹I from 51 wells that are used to monitor the ESRP aquifer. Six replicate samples and two blanks were collected as a measure of quality control. Concentrations of ¹²⁹I in the aquifer ranged from 0.0000013 ± 0.0000005 in a well near Kimama, Idaho to 1.02±0.04 pCi/L in a well south of INTEC, and concentrations generally decreased in wells near the INTEC from samples collected previously. The average concentration of ¹²⁹I in 15 wells sampled during four different sample periods decreased from 1.15 pCi/L in 1990-91 to 0.173 pCi/L in 2011-12. All but two wells within a 3-mile radius of the INTEC showed decreases in concentration, and all but one sample had concentrations less than the USEPA MCL. These decreases are attributed to the discontinuation of disposal of ¹²⁹I in wastewater after 1988 and to dilution and dispersion in the aquifer. The decreases around INTEC, in the wells where increases occurred between the 2003 and 2007 sample periods, were attributed to less recharge near INTEC either from less flow in the Big Lost River or from less local snowmelt and anthropogenic sources.

Wells sampled for the first time around the NRF had ¹²⁹I concentrations slightly higher than background concentrations in the ESRP aquifer. These concentrations are attributed to either seepage of unknown wastewater sources discharged at the Naval Reactors Facility or seepage from air emission deposits from INTEC, or both.

Although wells near the INTEC sampled in 2011–12 showed decreases in concentrations compared with previously collected data, some wells south and east of the Central Facilities Area, near the site boundary, and south of the INL showed slight increases. These slight increases are probably related to variable discharge rates of wastewater that eventually have moved to these well locations as a mass of water from a particular disposal period.

In 2012, the USGS collected samples from 25 zones in 11 wells equipped with multi-level monitoring systems to help define the vertical distribution of ¹²⁹I in the aquifer. Concentrations ranged from 0.000006 ± 0.000004 to 0.082 ± 0.003 pCi/L. Two new wells completed in 2012 showed variability of up to one order of magnitude of concentrations of ¹²⁹I among various zones. Two other wells showed a similar concentration of ¹²⁹I in all three zones sampled. Concentrations were well below the MCL in all zones.

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