

Concentrations of Selected Trace Metals, Common Ions,
Nutrients, and Radiological Analytes in Ground Water
for Selected Sites, Snake River Plain Aquifer, South of the
Idaho National Laboratory, Idaho



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Abstract

The state of Idaho, Department of Environmental Quality, Division of INL Oversight and Radiation Control (DEQ-INL) is charged with providing the citizens of Idaho with unbiased information concerning the impact of the INL on the state and its inhabitants. Tools utilized by the DEQ-INL to assess INL impacts and to verify conclusions reached by INL environmental monitoring include co-sampling and independent sampling to address specific questions. These samplings collectively can be used to address the question “Is the DEQ-INL monitoring in the right place?” to observe INL impacts down gradient in the Eastern Snake River Plain Aquifer south of the INL boundary.

Ground water samples from the Eastern Snake River Plain Aquifer were collected in 1997-1998 at 13 sites south of the INL, including six sites that were previously sampled for iodine-129 by the U.S. Geological Survey in 1991-1993. Samples from all sites were analyzed for tritium, iodine-129, gross alpha and beta radioactivity, gamma-emitting radionuclides, common ions, selected trace metals and nutrients. Selected sites were also sampled for chlorine-36. Analytical results were generally comparable to published ambient concentrations, and none of the constituents approached applicable drinking water maximum contaminant levels set by the U.S. Environmental Protection Agency.

Distribution of results for iodine-129, tritium, and chlorine-36 reflected three sources for these radionuclides for the samples collected; historical INL waste disposal, *in-situ* production within the aquifer, and infiltrating surface waters from the Wood River drainage. Surface waters include iodine-129, tritium, and chlorine-36 from cosmogenic and anthropogenic sources. The distribution of these isotopes in the aquifer suggests that ambient levels for these isotopes vary across the aquifer and are less in the central part of the aquifer ($<1 \times 10^{-6}$ pCi/L for iodine-129, $< 10 - 15$ pCi/L for tritium, and $>2 \times 10^{-4}$ pCi/L for chlorine-36). Two wells interpreted to have INL iodine-129 and tritium also yielded chlorine-36 concentrations indicative of INL impact.

An adaptation of the two-sample t-test for samples with different variances was used to compare iodine-129 results from the 1991-1993 samples collected by the US Geological Survey and the 1998 samples collected by DEQ-INL at six sites. At the 95 percent confidence level, samples from all sites differed from 1991-1993 to 1998; and for all but one, the 1998 concentration was less. The median for these sites decreased from 7.3×10^{-6} pCi/L in 1991-93 to 3.0×10^{-6} pCi/L in 1998. The greatest differences were noted between samples collected from Coffee Point well and Well #2.

Review of available information suggests that regional flow in the aquifer is to the south and west from the INL boundary. As flow appears to be more towards the Houghland well instead of the Coffee Point well and Well #2, monitoring should be focused more in this direction to capture the INL impacts reflected by environmental tracers such as iodine-129, tritium and chlorine-36.



Acknowledgements

The author would like to acknowledge the significant contributions that Dr DeWayne Cecil made to this effort, and to the author's understanding of the issues related to the use of long-lived anthropogenic radionuclides, as well as AMS methods for sample analysis.

Dr Cecil took the lead in bringing to publication the USGS report (Cecil, L.D., L.F. Hall, and J.R. Green, 2003) that includes results of the 1991-1993 iodine-129 sampling for sites visited in this study. He also provided technical review of this document.



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List of Acronyms

AMS	accelerator mass spectrometry
DEQ	Department of Environmental Quality
DEQ-INL	Division of INL Oversight and Radiation Control
ESP	Environmental Surveillance Program
INL	Idaho National Laboratory
INTEC	Idaho Nuclear Technology and Engineering Center
MV	Magic Valley
MCL	maximum contaminant level
µg/L	micrograms per liter
mg/L	milligrams per liter
NWQL	National Water Quality Laboratory
pCi/L	picocuries per liter
PRIME	Purdue Rare Isotope MEasurement
SMCL	Secondary maximum contaminant level
TRA	Test Reactor Area
USGS	United States Geological Survey



Introduction

Historical waste disposal practices at the Idaho National Laboratory (INL) have resulted in the presence of contaminants in the Eastern Snake River Plain Aquifer. These contaminants, iodine-129, chlorine-36, and tritium, as well as many other radioactive and chemical constituents, were contained in waste waters from nuclear reactor operations and spent nuclear fuel reprocessing, released to the environment via injection wells and infiltration ponds at INL facilities (Cecil and others, 1992; Beasley and others, 1993; Mann and Beasley, 1994a; Mann and Beasley, 1994b), and potentially, buried radioactive wastes. Historically, the distribution of tritium in the aquifer has been used as a benchmark for the extent of INL wastewater disposal impacts on the Eastern Snake River Plain Aquifer. However, with the passage of time, tritium is reaching the end of its usefulness for tracking the furthest extent of INL wastes in the Eastern Snake River Plain Aquifer due to its relatively short half-life (12.3 y), and its abundance from natural sources (Beasley and others, 1993).

Department of Environmental Quality, Division of INL Oversight and Radiation Control (DEQ-INL) provides Idaho citizens with unbiased information concerning INL impacts on the state by collecting environmental samples to verify conclusions reached by INL monitoring. These monitoring data, along with the supporting water quality and radiological samples collected concurrently, will be used to provide a basis for addressing, in general terms, the question of “Are we (the INL and DEQ-INL) monitoring in the right place?” to identify INL contaminants down gradient in the Eastern Snake River Plain Aquifer south of the INL boundary.

Background: Iodine-129 and Chlorine-36 in the Eastern Snake River Plain Aquifer

Iodine-129 and chlorine-36 both have very long half-lives (15.7 million years, and 0.301 million years respectively). In addition to having a cosmogenic source, in situ production under certain geologic conditions and as a result of nuclear weapons testing, both of these contaminants are also waste products from reprocessing spent nuclear fuel at the Idaho Nuclear Technology and Engineering Center (INTEC) (**Figure 1**). Chlorine-36 was also produced to some degree by reactor operations at the Reactor Technology Complex (RTC) formerly the Test Reactor Area (Cecil and others, 1992; Beasley and others, 1993; Mann and Beasley, 1994a, 1994b). Natural production of iodine-129 and chlorine-36 are discussed in Mann and Beasley, (1994a, 1994b); Beasley and others, (1993); Cecil, 2000, and Cecil and others (2003).

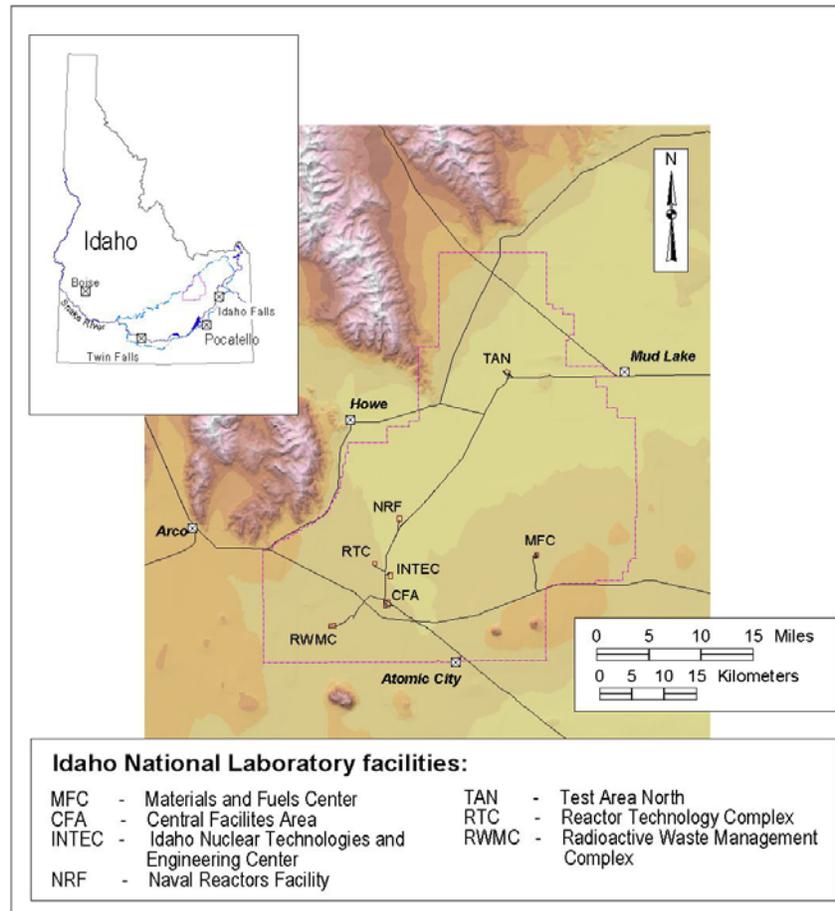


Figure 1. Location of the INL

Both iodine-129 and chlorine-36 travel conservatively, typically as negative ions (Rao, 1997; Beasley and others; 1998, Cecil, 2000). This is significant in that these negatively charged ions would tend to travel in the larger pore spaces, where water velocities are greatest. Recent work has suggested that although iodine-129 travels conservatively it appears to be slightly retarded relative to chlorine-36, possibly due to its affinity to organic materials (Beasley and others, 1998). Due to the long half lives of iodine-129 and chlorine-36 relative to tritium and the contrast between concentrations of iodine-129 and chlorine-36 from natural and anthropogenic sources (global fallout and INL waste disposal), these isotopes are potentially more valuable than tritium as a tracer for INL impacts to ground water when those impacts reach the far field.

Monitoring for iodine-129 in the aquifer beneath the INL began in 1977 by the U.S. Geological Survey (USGS). In subsequent years 1981, 1986, and 1990-91, the USGS conducted special sampling campaigns to investigate the distribution of this contaminant, and glean estimates of travel times in the aquifer based on this distribution. Twenty sites were sampled in 1977, 26 in 1981, and 35 in 1986 (Barraclough and others, 1982; Lewis and Jensen, 1985; Mann and others, 1988). Eighteen sites were sampled in each of the 1977, 1981, and 1986 sampling campaigns. The average iodine-129 concentration for these sites in 1977 was 4.0 pCi/L, with a maximum of



27 ± 1 . The average for 1981 sampling was 6.7 pCi/L , with a maximum of 41 ± 2 . Sample results for 1986 averaged 1.3 pCi/L , with a maximum of 3.6 ± 0.4 . Samples collected over this period were analyzed for iodine-129 by neutron activation by Radiological and Environmental Sciences Laboratory at the INL.

Mann and Beasley (1994b) sampled 51 wells in 1990-91 including locations up gradient of the injection well source at INTEC, and south of the INL southern boundary and utilized accelerator mass spectrometry (AMS), a methodology yielding a minimum detectable concentration (MDC) as much as six orders of magnitude lower than the neutron activation method used previously. Overall, sample results ranged from less than $0.6 \pm 0.2 \times 10^{-6} \text{ pCi/L}$ to $3.82 \pm 0.19 \text{ pCi/L}$. For the same 18 sites sampled in each of the previous sampling campaigns, the average concentration decreased to 0.81 pCi/L . Results from this sampling confirmed iodine-129 in aquifer waters beyond the INL's southern boundary (wells USGS 11 and 14 which are 6 and 12 km south of the boundary respectively).

By 1990, an estimated 0.56 to 1.18 curies of iodine-129 had been disposed of to the injection well and the percolation ponds at INTEC (solely to the injection well prior to 1983, and solely to the percolation pond following 1985). From 1987 until spent nuclear fuel reprocessing ceased in 1993, releases of iodine-129 from INTEC are identified as being released directly to the atmosphere instead of to liquid wastes. Discharged wastes were not analyzed for chlorine-36. However, based on an observed ratio of chlorine-36 to tritium in waste waters, an estimated seven curies of chlorine-36 was disposed of by INL activities while more than 31,000 curies of tritium was disposed of beneath the INL (Cecil, 2000; Bartholomay and others, 1997).

With the advent of the AMS method for iodine-129, and its much greater sensitivity and understanding of what bomb-related and natural production levels of iodine-129 should be, ambient concentrations could be estimated. Mann and Beasley (1994a) reviewed results for samples from the Eastern Snake River Plain Aquifer and tributaries, with values ranging from $1.0 \pm 0.4 \times 10^{-6} \text{ pCi/L}$ to $7.9 \pm 0.8 \times 10^{-6} \text{ pCi/L}$, a median of 3.0×10^{-6} , with 99 percent of values less than $8.2 \times 10^{-6} \text{ pCi/L}$, based on a subset of samples from 16 locations not likely to be impacted by the INL. Cecil and others (2003) incorporated sample results from an additional 32 locations representative of the Eastern Snake River Plain Aquifer to refine the Mann and Beasley (1994a) iodine-129 background estimate. Results for the total of 34 background samples ranged from $0.29 \pm 0.18 \times 10^{-6} \text{ pCi/L}$ to $10.0 \pm 0.4 \times 10^{-6} \text{ pCi/L}$, with a median of 3.3×10^{-6} and a background (defined by the 95th percentile value) of 5.4×10^{-6} (Cecil and others, 2003).

Chlorine-36 with its long half-life is also a permanent addition to the environment. The first analyses for chlorine-36 were conducted for water, snow, and soil samples collected in 1990. The purpose of these initial samples was to confirm that the isotope was released due to fuel reprocessing at the INL. Initial work focused on examining chlorine-36 releases to the atmosphere from INTEC (Cecil and others, 1992). Further investigations concluded that this nuclide would also be present in wastewater infiltrated or injected to the aquifer beneath the INL.



While the presence of chlorine-36 due to cosmogenic and anthropogenic sources was well known prior to 1990, no analyses were performed for this isotope either in waste streams or in environmental samples. Water monitoring in 1990-91 for chlorine-36 established an initial estimate of the distribution of this waste disposal product, as well as variability of chlorine-36 in the near environment not affected by INL operations (Beasley and others, 1993). Regular monitoring for this isotope has continued since then, including analysis of archived water samples (Cecil and others, 1998). A historical perspective on the development of waste disposal plumes beneath the INL and on chlorine-36 inputs from various sources (meteoric, *in-situ* production, bomb-pulse, and INL contributions) is documented in Cecil (2000).

Purpose and Scope

This sampling effort was designed to provide a follow up to the investigation reported by Cecil and others (2003) where samples were collected in 1991 and 1993 from selected aquifer wells down gradient of the INL. The further intent was to collect sufficient sample information to address the questions introduced previously in this report; “Is the DEQ-INL monitoring in the right place?” to observe INL impacts down gradient in the Eastern Snake River Plain Aquifer south of the INL boundary.

This question is addressed by:

- Presenting results for samples from thirteen selected sites sampled in 1997-1998;
- Discussing results for iodine-129, chlorine-36, and tritium;
- Drawing conclusions based on published values for expected ambient concentrations for these isotopes (Mann and Beasley, 1994a, 1994b; Mann and Low, 1994; Cecil and others, 1998; and Cecil, Knobel, and others, 2000); and
- Comparing selected iodine-129 results from this study with a subset of samples collected in 1991-1993 as a follow up to Mann and Beasley (1994a) and reported by Cecil and others (2003) for the purpose of establishing a background concentration for iodine-129 in the Eastern Snake River Plain Aquifer. The object of this comparison is to identify changes in iodine-129 concentrations over time for selected sites.

Data collection

Sample collection methods followed DEQ-INL and USGS protocols for sample collection and handling. The analyzing laboratories utilized standard methods for analysis.

Site Selection

Water samples were collected at 13 sites (**Figure 2**). Four sites were sampled by the USGS Magic Valley sampling program in 1997 (MV 40, 45, 47, 49) and the remaining nine were sampled by DEQ-INL, one in 1997 (Mindoka Water Supply) and eight in 1998 (Well #2, Coffee Point well, Crossroads well, Houghland well, and USGS 11, 14, 124, and 125). Sites sampled in



1998 included six locations previously sampled by Mann and Beasley during 1991-1993. Sites selected were beyond the INL boundary and along approximate aquifer flow paths south of the INL. The selected sites either showed historic INL contamination (USGS 11, 14, 124, and 125), or are characterized by typically low tritium (< 20 pCi/L) and low nitrate concentrations (< 1 mg/L), indicators that the sites are somewhat isolated from surface water impacts and INL contamination sources. One exception also sampled was MV 49, located along the northern margin of the Eastern Snake River Plain Aquifer.

Sample Collection

All the wells sampled were equipped with dedicated pumps. Prior to each sample collection, wells were pumped to remove standing water in the borehole and any associated plumbing such as the pressure tank and discharge line. During well purging, pH, specific conductance, and water temperature were monitored. After these parameters had stabilized and approximately three well-bore volumes had been pumped, samples were collected.

Where it was not possible to determine the wetted volume of the well bore (either the water level could not be measured and/or the well construction was not known with sufficient detail), the sample point or well was purged until the field parameters stabilized. This was accomplished in accordance with DEQ-INL standard procedures for collecting water samples. Samples from Magic Valley sites were collected by the USGS INL Project Office following USGS procedures. At the time of sampling, the pump for the Coffee Point well was not operating. However, an adjacent, large-volume storage tank did contain water. Samples were taken from the hydrant below this tank, after purging the sample point until field parameters had stabilized.

Field parameter measurements, along with well and sample collection information, are listed in **Table 1**. Final water temperatures ranged from 10.3 to 15.8°C with a mean of 13.6°C. Measurements of pH and specific conductance showed ranges of 7.71 to 8.27, with a mean of 8.04, and 309 to 449 $\mu\text{S}/\text{cm}$, with a mean of 364. These values are typical for ground water of the Eastern Snake River Plain Aquifer. It is interesting to note that while the mean water temperature was 13.6°C, the median value was 14.4°C. Three sites showed water temperatures about 30 percent colder than the most frequently observed value, while the highest temperature was only 10 percent greater than this mean.

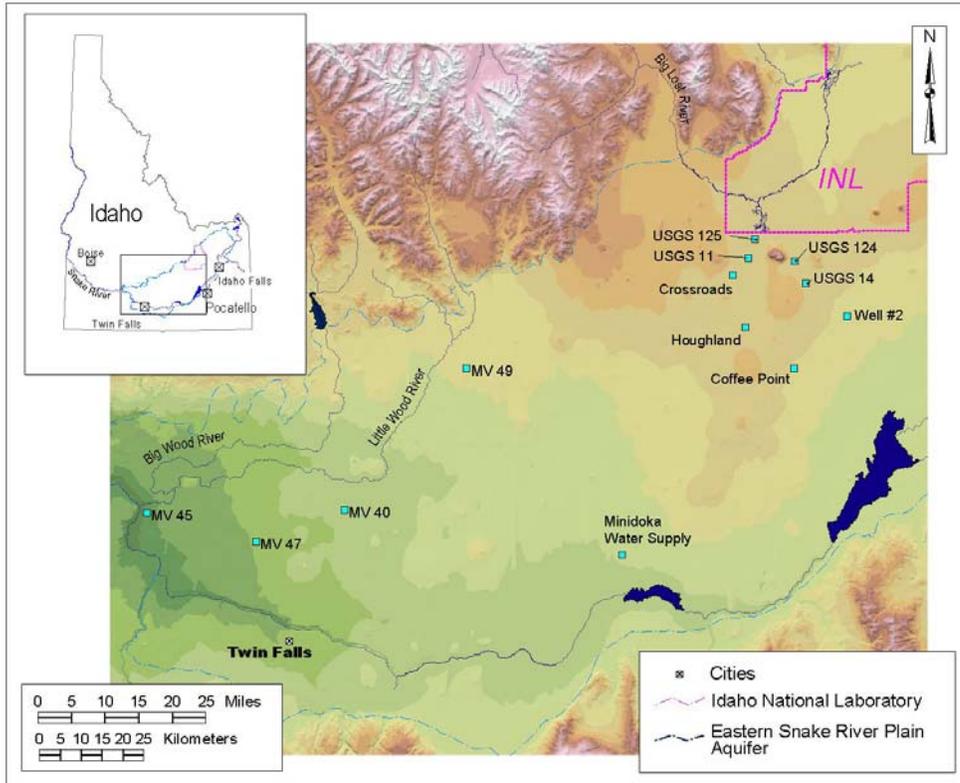


Figure 2. Ground water sites sampled 1997-98

Table 1. Field parameters for selected southern boundary samples, 1997-98.

Well Name	Sample Date	Temperature	pH	Conductivity
Coffee Point	6/18/98	14.4	8.23	319
Crossroads	6/23/98	10.5	8.27	309
Houghland	6/18/98	15.2	8.15	339
Minidoka Water Supply	8/5/97	15.5	7.71	449
USGS 11	10/7/98	11.6	8.09	355
USGS 14	10/7/98	14.4	8.12	389
USGS 124	10/8/97	13.2	8.04	369
USGS 125	10/7/98	12.3	8.05	376
Well #2	6/18/98	14.4	8.16	314
MV 40	8/25/97	14.5	8.01	374
MV 45	8/25/97	15.8	7.95	398
MV 47	8/25/97	14.8	7.97	376
MV 49	8/28/97	10.3	7.71	361



Chemical constituents

Samples collected for selected dissolved metals, dissolved nutrients, and common ions were analyzed using standard methods by the Idaho Bureau of Laboratories, Boise (IBL), and by the National Water Quality Laboratory (NWQL) for the Magic Valley sites. Samples for selected trace metals and nutrients were filtered in the field, while iodide analyses were performed on archived samples by the NWQL and filtered in the laboratory. A consistent set of trace metal, common ions, and nutrient analytical results were not available for all sites sampled in 1997-1998. Results from the most current sampling were included allowing the sites sampled to be compared with published background concentrations

Trace metals

Water samples were analyzed for selected dissolved trace metals, including arsenic, barium, cadmium, chromium, copper, iron, lead, manganese, nickel, silver, zinc selenium, and mercury (Table 2).

Concentrations of arsenic in the Eastern Snake River Plain Aquifer from previous water quality studies suggest a typical range of 2 to 3 µg/L (Knobel and others, 1992). Samples from the Magic Valley (MV) sites ranged from <1 to 2.52 µg/L. All samples from the remaining sites were less than the detection level for the IBL, 10 µg/L. The maximum contaminant level (MCL) for arsenic is 50 µg/L.

Table 2. Dissolved trace metal concentrations¹ for selected southern boundary samples, 1997-98. Concentrations are expressed in µg/L.

Sample Location	Sample Date	Concentration						
		Arsenic	Barium	Cadmium	Chromium	Copper	Iron	Lead
Coffee Point	6/18/98	<10	17.0	<1	3	<10	<10	<5
Crossroads	6/23/98	<10	35.0	<1	3	<10	25	<5
Houghland	6/18/98	<10	12.0	<1	4	<10	<10	<5
Minidoka Water Supply	8/5/97	<10	47.0	<1	2	<10	<10	<5
USGS 11	10/7/98	<10 ²	48.0	<1 ²	4	<10 ²	<10 ²	<5
USGS 14	10/7/98	<10 ²	19.0	<1 ²	5	<10 ²	<10 ²	<5
USGS 124	10/8/97	<10 ²	28.0	<1 ²	6	<10 ²	54	<5
USGS 125	10/7/98	³	33.0	³	5	³	³	<5
Well #2	6/18/98	<10	20.0	<1	3	<10	45	<5
MV 40	8/25/97	2.52	18.9	<1	<5	<10	7.2	33.1
MV 45	8/25/97	2.07	23.2	<1	<5	<10	<3	<10
MV 47	8/25/97	2.33	18.3	<1	<5	<10	<3	31.5
MV 49	8/28/97	<1	49.2	<1	<5	<10	71.6	<10

¹ A "<" indicates a result below the Minimum Reportable Value

² Analytical results are from samples collected 10/6/97

³ Not analyzed for



Table 2 continued. Dissolved trace metal concentrations¹ for selected southern boundary samples, 1997-98. Concentrations are expressed in µg/L.

Sample Location	Sample Date	Concentration					
		Manganese	Nickel	Silver	Zinc	Selenium	Mercury
Coffee Point	6/18/98	<1	<5	<1	17	<5	<0
Crossroads	6/23/98	<1	<5	<1	30	<5	<0
Houghland	6/18/98	<1	<5	<1	28	<5	<0
Minidoka Water Supply	8/5/97	<1	<5	<1	13	<5	<0.5
USGS 11	10/7/98	<1	<5 ²	<1 ²	76	<5 ²	<0.5 ²
USGS 14	10/7/98	<1	<5 ²	<1 ²	39	<5 ²	<0.5 ²
USGS 124	10/8/97	8	<5 ²	<1 ²	4	<5 ²	<0.5 ²
USGS 125	10/7/98	6	₃	₃	<2	₃	₃
Well #2	6/18/98	1	<5	<1	47	<5	<0
MV 40	8/25/97	<1	<10	<1	6.6	<1	<0.1
MV 45	8/25/97	1.6	<10	<1	<3	<1	<0.1
MV 47	8/25/97	<1	<10	<1	7.1	<1	<0.1
MV 49	8/28/97	9.1	<10	<1	54.6	<1	<0.1

¹ A "<" indicates a result below the Minimum Reportable Value
² Analytical results are from samples collected 10/6/97
³ Not analyzed for

Concentrations of barium ranged from 12 to 49.2 µg/L with a median of 23.2. Ambient concentrations based on previous studies suggest a natural range of 50 to 70 µg/L (Knobel and others, 1992). Barium concentrations were slightly higher for wells USGS 11, 125, Crossroads Well, Minidoka water supply, and MV 49 than for others. The MCL for barium is 2000 µg/L. Barium was identified in liquid wastes from several INL facilities at about 10 percent of the MCL (Frederick and others, 1998) and was also elevated in some INL wells monitored by the DEQ-INL (Hall, 2004).

Chromium concentrations ranged from 2 to 6 µg/L, and were all less than the reporting level of 5 µg/L for MV sites. The median value for all sites was 3 µg/L. Ambient concentrations based on DEQ-INL monitoring ranged from about 2 to 7 µg/L with a median of 3 (Hall, 2004). The MCL for chromium was 100 µg/L. Chromium was elevated in some INL wells monitored by the DEQ-INL and exceeded the MCL at one site on the INL.

Concentrations for iron ranged from less than the reporting level (3 µg/L for Magic Valley sites and 10 µg/L for other sites) to 71.6 µg/L, with most sites less than the reporting level. Concentrations for sites MV 49 and 40 were 71.6 and 7.2 µg/L with the median concentration of 5 for all MV sites. The median value for the remaining sites was less than the 10 µg/L reporting level. The secondary MCL (SMCL) for iron was 300 µg/L. Knobel and others (1999) reported concentrations ranging from 4 to 210 µg/L, with a median of 16.



Lead concentrations for the aquifer by previous studies ranged from 1 to 4 µg/L (Knobel and others, 1992). Most samples showed concentrations less than the reporting level, 5 µg/L for the MV sites and 10 µg/L for the remaining locations. Two Magic Valley sites showed concentrations of 32 to 33 µg/L. Lead is regulated at an action level of 15 µg/L.

Manganese was found in small concentrations in just a few samples. Reported values ranged from less than the reporting level to 9.1 µg/L with the median concentration reported for all sites of less than the reporting level of 1 µg/L. The SMCL for manganese was 50 µg/L. Manganese was identified in some liquid wastes at INL, but not typically detected in DEQ-INL monitoring. Knobel and others (1999) reported concentrations ranging from less than 1 to 83 µg/L, with a median of 3.

Zinc concentrations ranged from less than 2 to 82 µg/L with a median of 17 for all sites. The SMCL for zinc was 5000 µg/L. Knobel and others (1999) reported concentrations ranging from less than 3 to 420 µg/L, with a median of 11.

All samples for these analytes were less than the reportable value; <1 µg/L for cadmium, <10 µg/L for copper, <5 µg/L and <10 µg/L for nickel, <1 µg/L for silver, <1 µg/L and <5 µg/L for selenium, and <0.1 µg/L and <0.5 µg/L for mercury. Ambient concentrations for these constituents are expected to be less than the reporting level. Some of these analytes are identified in INL wastes, but are not commonly present above minimum detectable concentrations.

Common ion and dissolved nutrients

Table 3 presents comparable common ion and nutrient results. For natural ground waters, calcium, magnesium, sodium, potassium, chloride, sulfate, total alkalinity, and total nitrate plus nitrite, along with silica typically comprised as much as 99 percent of the dissolved constituents. These constituents are referred to as the major ions or common ions. For MV sites, a more complete list of common ions and nutrients were sampled for prior to 1995. These data are included for comparisons.

Calcium concentrations ranged from 30 to 44.6 mg/L with a median of 36.6. While calcium was not identified as a major constituent of INL waste waters, ground water concentrations of calcium down gradient of INL facilities that disposed of large quantities of waste water were about 30 percent higher than ambient levels up gradient from the INL. Knobel and others (1999) reported concentrations ranging from 5.4 to 88 mg/L, with a median of 43.



Table 3 Common ions and nutrient concentrations¹ for selected southern boundary samples, 1997-98. Concentrations are expressed in mg/L.

Sample Location	Sample Date	Concentration						
		Calcium	Magnesium	Sodium	Potassium	Flouride	Chloride	Sulfate
Coffee Point	6/18/98	31.0	13.0	13.0	2.9	0.59	13.6	15.4
Crossroads	6/23/98	43.0	14.5	7.2	2.1	0.23	9.7	20.5
Houghland	6/18/98	31.2	15.6	12.0	3.2	0.53	14.6	18.8
Minidoka Water Supply	8/5/97	44.6	16.4	19.0	3.3	0.69	28.1	36.4
USGS 11	10/7/98	40.9	14.8	8.4	2.2	0.26	11.3	22.9
USGS 14	10/7/98	36.6	15.9	16.0	2.8	0.91	21.9	22.9
USGS 124	10/8/97	38.7	16.9	9.6	2.4	0.37	15.0	22.5
USGS 125	10/7/98	38.9	16.0	12.0	2.9	0.27	14.0	25.2
Well #2	6/18/98	30.0	12.7	13.0	2.9	0.68	12.8	14.2
MV 40	8/25/97	31.0	15.0	18.0	2.8 ³	0.4 ³	10 ³	23 ³
MV 45	8/25/97	34.0	17.0	18.0	3.8 ³	0.4 ³	11 ³	24 ³
MV 47	8/25/97	32.0	16.0	18.0	3.4 ³	0.6 ³	15 ³	32 ³
MV 49	8/28/97	45.0	13.0	7.0	2.1 ³	0.2 ³	3.1 ³	15 ³

¹ A "<" Indicates a result below the Minimum Reportable Value
² Analytical results are from samples collected 10/06/97
³ Analytical results are from previous samples, MV 40 collected 8/10/94; MV 45 collected 06/20/95; MV 49 collected 8/16/94
⁴ Sample was not analyzed due to exceeding holding time on receipt at lab
⁵ Analysis not requested.

Table 3 continued. Common ions and nutrient concentrations¹ for selected southern boundary samples, 1997-98. Concentrations are expressed in mg/L.

Sample Location	Sample Date	Concentration						
		Silica	Bicarbonate Alkalinity	Carbonate Alkalinity	Total Dissolved Solids	Total Nitrate Nitrite	Total Phosphorus	Dissolved Iodine
Coffee Point	6/18/98	33.3	120	1	⁴	0.818	<0.05	0.008
Crossroads	6/23/98	23.6	131	9	231	0.740	<0.05	0.001
Houghland	6/18/98	32.7	117	10	⁴	0.749	<0.05	0.016
Minidoka Water Supply	8/5/97	29.7	137	<1	289	0.866	<0.05	0.003
USGS 11	10/7/98	21.3 ²	139	6 ²	211 ²	0.649	<0.05	0.006
USGS 14	10/7/98	29.5 ²	135	²	234 ²	1.130	<0.05	0.005
USGS 124	10/8/97	24.8 ²	141	3 ²	221 ²	0.786	<0.05	0.003
USGS 125	10/7/98	⁵	146	⁵	⁵	0.595	<0.05	0.003
Well #2	6/18/98	33.6	112	11	⁴	0.795	<0.05	0.011
MV 40	8/25/97	32.0	125 ³	⁵	⁵	0.888	0.011	0.001
MV 45	8/25/97	32.0	137 ³	⁵	⁵	1.257	0.015	0.003
MV 47	8/25/97	32.0	130 ³	⁵	⁵	1.183	0.021	0.009
MV 49	8/28/97	25.0	169 ³	⁵	⁵	1.785	0.020	0.009

¹ A "<" Indicates a result below the Minimum Reportable Value
² Analytical results are from samples collected 10/06/97
³ Analytical results are from previous samples, MV 40 collected 8/10/94; MV 45 collected 06/20/95; MV 49 collected 8/16/94
⁴ Sample was not analyzed due to exceeding holding time on receipt at lab
⁵ Analysis not requested.



Magnesium concentrations ranged from 12.7 to 17.4 mg/L with a median of 15.5. As with calcium, magnesium concentrations were elevated down gradient of INL facilities and in some wells monitored by the DEQ-INL. A range of less than 1 to 23 mg/L with a median of was reported by Knobel and others (1999).

Sodium concentrations ranged from 7.8 to 19 mg/L with a median of 13. Sodium was a significant constituent in INL chemical wastes disposed of to the Eastern Snake River Plain Aquifer, and has an identifiable waste plume beneath the INL. Maximum concentrations for INL monitoring wells were as much as four to five times ambient concentrations up gradient from the INL. Knobel and others (1999) reported concentrations ranging from 5.4 to 47 mg/L, with a median of 14.

Potassium concentrations ranged from 2.1 to 3.8 mg/L with a median of 2.9. Potassium was identified in INL waste waters; however, aquifer concentrations were only slightly greater than ambient concentrations up gradient from the INL.

Fluoride concentrations ranged from 0.20 to 0.91 mg/L with a median of 0.40. Effluent concentrations for fluoride were typically only slightly higher than ambient ground water concentrations. Knobel and others (1992) reported a background range of 0.4 to 0.5 mg/L for the aquifer. Variations in fluoride concentrations may be related to ground water flow paths in the aquifer (Hall, 2000).

Chloride concentrations ranged from 3.1 to 28.1 mg/L with a median of 13.8. Chloride was another major constituent of liquid wastes, and had an identifiable plume in the aquifer beneath the INL. Knobel and others (1999) reported concentrations ranging from 2.6 to 120 mg/L, with a median of 16.

Sulfate concentrations ranged from 14.2 to 36.4 mg/L with a median of 22.9. Sulfate was a major wastewater constituent, with total mass disposed typically only exceeded by sodium and nitrate wastes for INL facilities. Sulfate also varied by a factor of three or four among up gradient, DEQ-INL water monitoring sites (INEEL OP, 2000). Knobel and others (1999) reported concentrations ranging from 2.0 to 200 mg/L, with a median of 24.

Silica concentrations ranged from 21.3 to 33.6 mg/L with a median of 31.7. Silica was a natural constituent of ground water and second only to oxygen in abundance in the earth's crust. Silica was present in natural ground water as a silicon-tetrahedron (SiO_4). Knobel and others (1999) reported concentrations from 10 to 48 mg/L with a median of 26.

Alkalinity is defined as the capacity for waters to react with and neutralize acids. This capacity is typically reported as an equivalent concentration of calcium carbonate, or a combination of bicarbonate and carbonate ions. Total alkalinity as bicarbonate alkalinity ranged from 112 to 169 mg/L with a median of 134.5. The bicarbonate ion (HCO_3) was the most abundant anion for most waters from the Eastern Snake River Plain Aquifer. The relative concentrations of bicarbonate and carbonate alkalinity for a sample were dependant on the pH of the waters



sampled, with the bicarbonate ion dominant at the range of pH's observed for the aquifer. Total alkalinity as bicarbonate alkalinity ranged from 106 to 276 mg/L with a mean of 144.9 for selected INL wells sampled by the USGS (Knobel and others, 1999), and from 99 to 261 mg/L with a mean of 169 for 55 sites sampled by the USGS in the Magic Valley region (Bartholomay and others, 1996; Bartholomay and others, 1995; Bartholomay and others, 1994).

Analytical results for total dissolved solids were available for five of the sites sampled, with concentrations ranging from 211 to 289 mg/L, with a median of 216, consistent with DEQ-INL historic aquifer sampling (Hall, 2004).

Nutrient concentrations (total nitrate plus nitrite and total phosphorus) ranged from 0.595 to 1.785 mg/L with a median of 0.818 for total nitrate plus nitrite, and from less than the detection level of 0.05 mg/L for total phosphorus samples analyzed by the IBL and 0.011 to 0.021 with a median of 0.015 for total phosphorus samples analyzed by NWQL. Nitrate is a significant component of INL waste waters, and total nitrate plus nitrate levels in specific INL monitoring wells are as much as 4 mg/L. Phosphorus is also a common waste constituent, but disposed of in smaller quantities than nitrate at the INL.

Iodine is present in the environment at trace levels. The concentration of iodine in geologic materials is typically very small. However, iodine tends to bind with organic materials such as decaying vegetation and thus can be concentrated in organic-rich soils (Rao, 1997). Analyses for dissolved iodine (iodide, I⁻) were obtained from archived samples and completed by NWQL for all sites. Iodide concentration ranged from 0.001 to 0.016 mg/L, with a median of 0.005. Mann and Beasley (1994b) assumed that concentrations of iodine in the aquifer were generally less than the analytical method detection limit of 0.001 mg/L. Iodine concentrations reported by Cecil and others (2003) ranged from 0.001 to 0.004 mg/L, with a median of 0.002.

Radiological Constituents

Gross alpha, gross beta, gamma spectroscopy, and tritium analyses were completed by Idaho State University Environmental Monitoring Laboratory (ISU-EML) (**Tables 4 and 5**). Analyses for iodine-129 and chlorine-36 utilized AMS (**Tables 6 and 7**) and were conducted by the PRIME (Purdue Rare Isotope MEasurement) Laboratory, Purdue University, West Lafayette, Indiana.

Analyses for gross alpha-particle radioactivity ranged from -2.3 ± 1.7 to 6.5 ± 1.8 pCi/L, with seven of thirteen samples showing detectable alpha radioactivity. The mean alpha concentration was 2.1 pCi/L. The MCL for gross alpha radioactivity in water from all sources is 15 pCi/L. Ambient concentrations for gross alpha radioactivity are typically less than 3 pCi/L, (Knobel and others, 1992).



Table 4. Gross alpha, beta, and gamma for selected southern boundary samples, 1997-98. Concentrations are expressed in pCi/L.

Location Name	Sample Date	Gross alpha		Gross Beta		Cesium-137		Potassium-40-	
		Concentration	±2SD	Concentration	±2SD	Concentration	±2SD	Concentration	±2SD
Coffee Point	6/18/98	6.5 ³	1.8	2.8 ³	0.8	3.6	3.1	19.8	55.8
Crossroads ²	6/23/98	3.6 ³	1.6	1.8 ³	0.7	2.7	2.5	-2.3	56.8
Houghland ^{1,2}	6/18/98	3.8 ³	1.5	1.6 ³	0.7	1.5	3.2	53.3	70.4
Minidoka Water Supply	11/04/97	2.7	5.2	4.3	3.1	0.6	3.0	54.0	68.0
MV 40	8/25/97	1.6	2.6	3.4 ³	1.3	-1.6	2.2	-21.0	46.0
MV 45	8/25/97	-0.8	2.8	3.3 ³	1.3	0.8	3.3	-16.4	63.4
MV 47	8/27/97	4.4 ³	2.7	2.4 ³	1.3	-1.8	2.8	-17.0	50.5
MV 49	8/27/97	-0.9	1.5	1.8	1.2	-1.6	2.7	23.1	65.4
USGS 11	10/07/98	2.4 ³	1.4	1.0	0.8	-0.7	2.5	11.2	68.3
USGS 14	10/07/98	3.6 ³	1.5	0.8	0.8	-0.5	3.3	102.7	74.4
USGS 124	10/08/98	-2.3	1.7	0.4	0.8	1.2	2.2	14.4	74.9
USGS 125	10/08/98	-2.1	1.9	1.3	0.9	0.7	2.3	14.7	67.9
Well #2	6/18/98	4.9 ³	1.4	1.2	0.8	0.1	2.7	-32.5	69.9

¹ Result is the average of samples split in the laboratory. ¹ Result is the average of samples split in the laboratory.

² Result is the average of multiple recounts of the same sample in the laboratory

³ Result exceeded sample-specific MDC (approximately 3-4 pCi/L for both gross alpha and gross beta).

Table 5. ESP sampling results for tritium concentrations at selected southern boundary sites, 1997-98. Concentrations are expressed in pCi/L.

Location Name	Sample Date	Concentration	±2SD
Coffee Point	6/18/98	1	7
Crossroads	6/23/98	24 ¹	7
Houghland	6/18/98	9	7
Minidoka Water Supply	8/05/97	1	7
MV 40	8/25/98	58 ¹	6
MV 45	8/25/98	48 ¹	5
MV 47	8/27/98	271 ¹	9
MV 49	8/28/98	57 ¹	8
USGS 11	10/07/98	126 ¹	8
USGS 14	10/07/98	7	4
USGS 124	10/08/98	12	8
USGS 125	10/07/98	16 ¹	7
Well #2	6/18/98	8	7

¹ Result exceeded sample-specific MDC (approximately 6-15) for tritium by the following electrolytic enhancement.



Analyses for gross beta-particle radioactivity ranged from 0.4 ± 0.8 to 4.3 ± 3.1 pCi/L, with six of thirteen samples showing detectable gross beta radioactivity. The mean beta concentration was 2.0 pCi/L. Ambient concentrations for gross beta radioactivity are typically less than 7 pCi/L (Knobel and others, 1992). The MCL for gross beta radioactivity is based on the combined exposure of 4 millirem per year for all beta-emitting and gamma-emitting radionuclides present in drinking water. Specific MCLs for beta-emitting radionuclides tritium and strontium-90 are 20,000 and 8 pCi/L.

Samples were analyzed for gamma-emitting radionuclides by gamma spectroscopy. No gamma-emitting radionuclides were detected in samples. Specific detection levels for cesium-137 and potassium-40 are about 6 and 70 pCi/L., concentrations were estimated by the ISU-EML based on the gamma-spectrum region of interest for cesium-137 and potassium-40.

Tritium was analyzed using a base electrolysis procedure for enriching tritium in the sample following standard methods described in HASL 300 (US DOE, 1992), and liquid scintillation counting. Results ranged from 1 ± 7 to 271 ± 9 pCi/L with a minimum detectable concentration of about 12-15 pCi/L. Knobel and others (1992) suggest a background for tritium in the Eastern Snake River Plain Aquifer of 0 to 40 pCi/L. Rupart (1997) suggests that aquifer waters with tritium concentrations greater than 4.5 pCi/L have been recharged prior to the onset of atmospheric atomic bomb testing. Two sample sites, USGS 124 and 125 show tritium levels indicative of INL waste tritium. Another two sites, USGS 11 and USGS 14, show tritium levels just greater than the background range of Knobel and others (1992). Based on their location, just south of the INL and only a couple of miles from sites showing obvious INL impacts, lends the conclusion that the tritium observed at USGS 11 and 14 is likely from the INL. Another site, MV 49 also shows tritium at the same levels as USGS 11 and 14. However, due to nitrate concentrations indicative of surface water impacts, a tritium concentration reflective of recent connection with surface waters, and based on DEQ-INL monitoring, the tritium for this site is concluded to not be of INL origin. The remaining sites show results less than detection or just barely above detection, indicative of very limited surface water influences and older ground water.

AMS Analyses

Results for analyses by AMS are expressed in terms of an isotopic ratio of the target isotope to stable isotope(s), (i.e. $^{129}\text{I}/^{127}\text{I}$ or $^{36}\text{Cl}/^{35,37}\text{Cl}$) for that specific sample. That ratio is used, along with the mass of sample, mass of iodine or chloride carrier added, to calculate the number of atoms per liter and activity-concentration of the target isotope.

Samples from 13 sites were submitted to the PRIME Laboratory, Purdue University, for iodine-129 analysis by AMS (**Table 6**). From the laboratory results, and along with the concentration of iodine in the sample, the number of atoms per liter iodine-129 and the pCi/L concentration of iodine-129 for that water sample can be calculated. Atoms per liter concentrations ranged from $4.2 \pm 3.5 \times 10^6$ to $20000 \pm 780 \times 10^6$. Pre-anthropogenic levels of iodine-129 range from 10^3 to



10^5 atoms per liter (Rao, 1997, Fabryka-Martin and others, 1987). Background concentrations for iodine-129 for Eastern Snake River Plain Aquifer sites range from $11.7 \pm 4.9 \times 10^6$ to $265 \pm 10.2 \times 10^6$ atoms per liter (Cecil and others, 2003).

Table 6. ESP sampling results for iodine-129 by AMS analysis at selected southern boundary sites, 1997-98.

Well	Sample Date	Atoms Iodine-129 per L		Iodine-129 (pCi/L)	
		$(\times 10^{-6})$		$(\times 10^{-6})$	
		Concentration	± 1 SD	Concentration	± 1 SD
Coffee Point	6/18/98	4.2	3.5	0.16	0.013
Crossroads	6/23/98	20.0	0.69	0.76	0.026
Houghland	6/18/98	140	5.8	5.2	0.19
Minidoka Water Supply	8/5/97	28.0	1.3	1.1	0.47
USGS 011	10/7/98	160	4.7	6.1	0.18
USGS 014	10/7/98	490	32.0	19.0	1.2
USGS 124	10/8/98	20000	780	740	30
USGS 125	10/7/98	810	250	31.0	9.3
Well #2	6/18/98	5.2	0.30	0.20	0.012
MV 40	8/25/97	130	5.5	4.9	0.21
MV 45	8/25/97	130	4.7	4.9	0.18
MV 47	8/27/97	94.0	3.6	3.6	0.14
MV 49	8/28/97	47.0	1.8	1.8	0.068

Concentrations of iodine-129 ranged from $0.16 \pm 0.013 \times 10^{-6}$ to $740 \pm 30 \times 10^{-6}$ pCi/L. Cecil and others (2003) established an estimated background concentration for the Eastern Snake River Plain Aquifer of 5.4×10^{-6} pCi/L, with a 95 percent confidence interval of $5.2 - 10.0 \times 10^{-6}$ pCi/L. Iodine-129 decays primarily by release of a beta-particle, and thus the associated MCL is based on the activity needed to exceed the 4-millirem-per year allowable limit for gross-beta radioactivity. This activity-concentration is 1 pCi/L (US Environmental Protection Agency)

Samples from four sites were submitted to PRIME Lab for AMS analysis of the ratio of chlorine-36 to stable chloride ($^{36}\text{Cl}/^{35,37}\text{Cl}$) (Table 7). The four sites had not been sampled for chlorine-36 previously. Atoms per liter concentrations ranged from $57.2 \pm 2.17 \times 10^6$ to $261 \pm 7.94 \times 10^6$. Cecil (2000) concluded that atoms per liter concentrations of chlorine-36 for Eastern Snake River Plain surface water or ground water between 100×10^6 and 1000×10^6 atoms may be attributed to a combination of factors including re-suspension of weapons-test fallout, concentration of chlorine-36 by evapotranspiration, and historic INL waste disposal operations. Concentrations greater than 1000×10^6 atoms per liter for the surface water and ground water of the Eastern Snake River Plain are directly attributable to the INL.



Table 7. ESP sampling results for chlorine-36 by AMS analysis at selected southern boundary sites, 1997-98.

Well	Sample Date	Atoms chlorine-36 per L		Chlorine-36 (pCi/L)	
		(x10 ⁶)		(x10 ⁻⁶)	
		Concentration	± 1 SD	Concentration	± 1 SD
Coffee Point	6/18/98	58	2.3	110	4.6
Crossroads	6/23/98	190	6.6	380	13.0
Houghland	6/18/98	260	7.9	520	16.0
Well #2	6/18/98	57	2.2	110	4.3

Activity-concentrations for chlorine-36 ranged from $113 \pm 4.29 \times 10^{-6}$ to $515 \pm 15.7 \times 10^{-6}$ pCi/L. Atoms per liter concentrations reported by Cecil (2000) as attributable to weapons-test fallout, INL impacts, and concentration by evapotranspiration correspond to approximately 200 to 2000 $\times 10^{-6}$ pCi/L, with concentrations greater than 2000 $\times 10^{-6}$ pCi/L attributable only to INL, based on typical chloride concentrations for ground water. Chlorine-36 decays primarily by release of a beta-particle, and thus the associated MCL is based on the activity needed to exceed the 4-millirem-per year allowable limit for gross-beta radioactivity. The corresponding activity-concentration at this MCL is 700 pCi/L.

Discussion of Results

Iodine-129 concentrations for selected sites presented in **Figure 3** show higher concentrations near the INL, generally decreasing to the south. Also shown in **Figure 3** is the approximate direction of ground water flow in the aquifer, based on Lindholm and others (1988).

Concentrations are highest for well USGS 124, just south of the INL and to the east of Big Southern Butte, decreasing by a factor

of about 40 towards USGS 14, a distance of approximately 5 km. Concentrations for Coffee Point well and Well #2 wells are more than four orders of magnitude lower than USGS 124. Cecil and others (2003) report iodine-129 background as $\leq 5.4 \times 10^{-6}$ pCi/L for the Eastern Snake River Plain Aquifer. Within this context, the results from samples near the Wood River, and results from samples in the center portion of the aquifer less than approximately 5×10^{-6} pCi/L are not likely to contain INL iodine-129, whereas wells USGS 124, 125, 11, and 14 likely do. The sample from the Houghland well is near the upper range for background, and might have INL iodine-129.

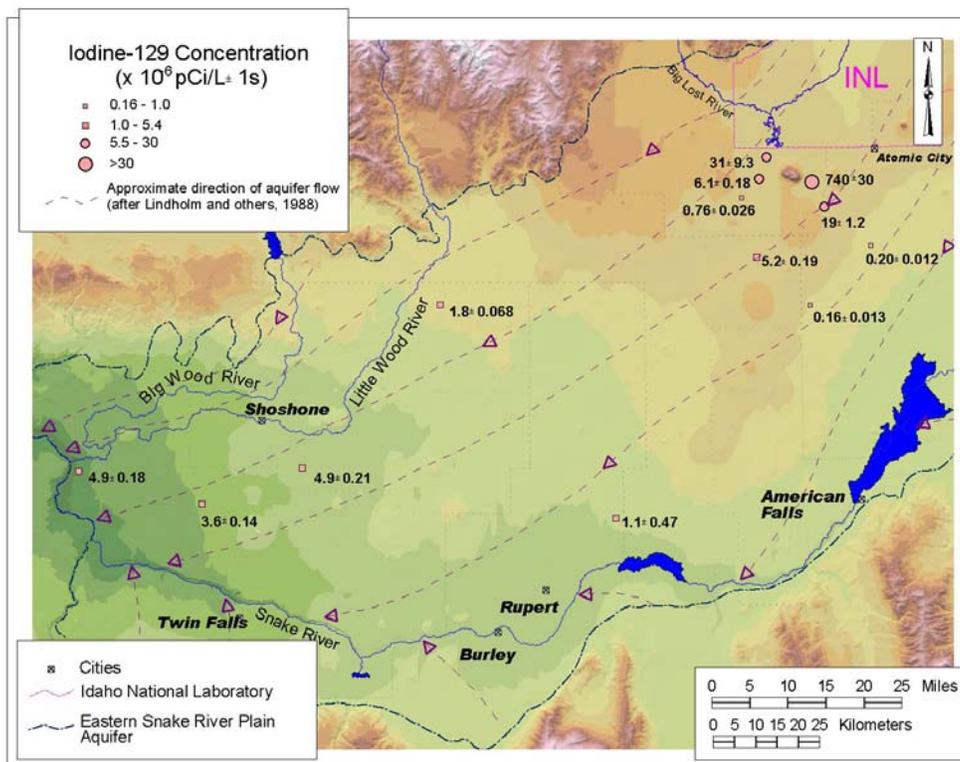


Figure 3. Iodine-129 concentrations

A limited number of samples were collected for chlorine-36 analysis, as previous samples for this analyte have not been collected from this subset of sites, Crossroads well, Houghland well, Coffee Point well, and Well #2. Results are presented in **Figure 4** along with the general direction of aquifer flow. Concentrations greater than 100×10^6 atoms per liter are indicative of anthropogenic impacts, including historic INL waste disposal (Cecil (2000)). This is equivalent to approximately 200×10^{-6} pCi/L chlorine-36. Based on these values, the Houghland well potentially shows indications of anthropogenic impacts, possibly from INL. This agrees with iodine-129 results where the Houghland well exceeded estimated ambient conditions for the portion of the Eastern Snake River Plain Aquifer down gradient from the INL

Tritium concentrations for selected sites and general direction of aquifer flow are presented in **Figure 5**. The distribution of tritium for these sites reflects that of iodine-129, with the highest concentration at well USGS 124, which decreases to the south and east from that point. The tritium concentration at this site obviously resulted from INL waste disposal. Mann and Low (1994) and Rupart (1997) addressed distribution of tritium in the Eastern Snake River Plain Aquifer. Mann and Low examined concentrations from selected springs and streams, devising a scheme to categorize the streams and springs by the relative amount of recent recharge, as judged by tritium concentrations.

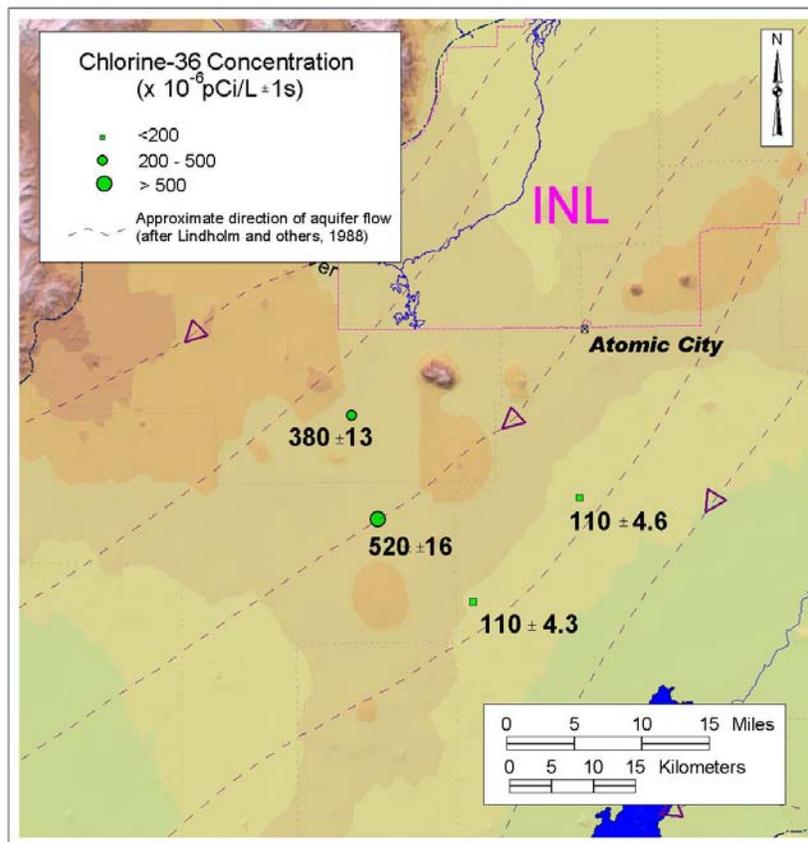


Figure 4. Chlorine-36 concentrations

Rupart utilized tritium concentrations to support conclusions on source of nitrates in ground water. Collectively, they agree that tritium concentrations from about 50 to 70 pCi/L represent recently recharged ground water for that time period. This assumption for a ground water sample would be supported by an accompanying total nitrate plus nitrite concentration greater than about 1 mg/L (Rupart, 1997) indicative of human impact. Natural production of tritium due to cosmic-ray interaction in the atmosphere is estimated to be less than 16 pCi/L (<5 tritium units) (Lehman and others, 1993). Thus ground water that recharged prior to 1952, the start of atmospheric bomb-testing, should have likely to have tritium at or below detectable levels. For tritium measurements from ISU-EML, this detection level is 10-15 pCi/L.

The distribution of tritium shown in **Figure 5** suggests a mixing of recent recharge and older ground waters for MV sites along the Wood River drainage. This is supported by total nitrate concentrations from these sites. The low-levels of tritium reported for samples in the central portion of the aquifer would be consistent with older ground water with little to no influence from recent surface waters. The sites to the west of Big Southern Butte show concentrations indicative of some impacts; either infiltrating waters from the Big Lost River or INL waste disposal, or a mixture of both. In light of iodine-129 concentrations from the previous figure, wells USGS 125, and 11 likely show INL impacts and Crossroads well may just be showing the



impact of infiltrating surface waters (based on tritium, and possibly chlorine-36, measurements). Conclusions from Rupert (1997), who also considered tritium results from Crossroads well and Houghland well, are that both include water that has been recharged recently (since 1952). These waters could be from either the nearby closed drainage basins on the Eastern Snake River Plain that fill with water in wet years, or infiltration from the Big Lost River.

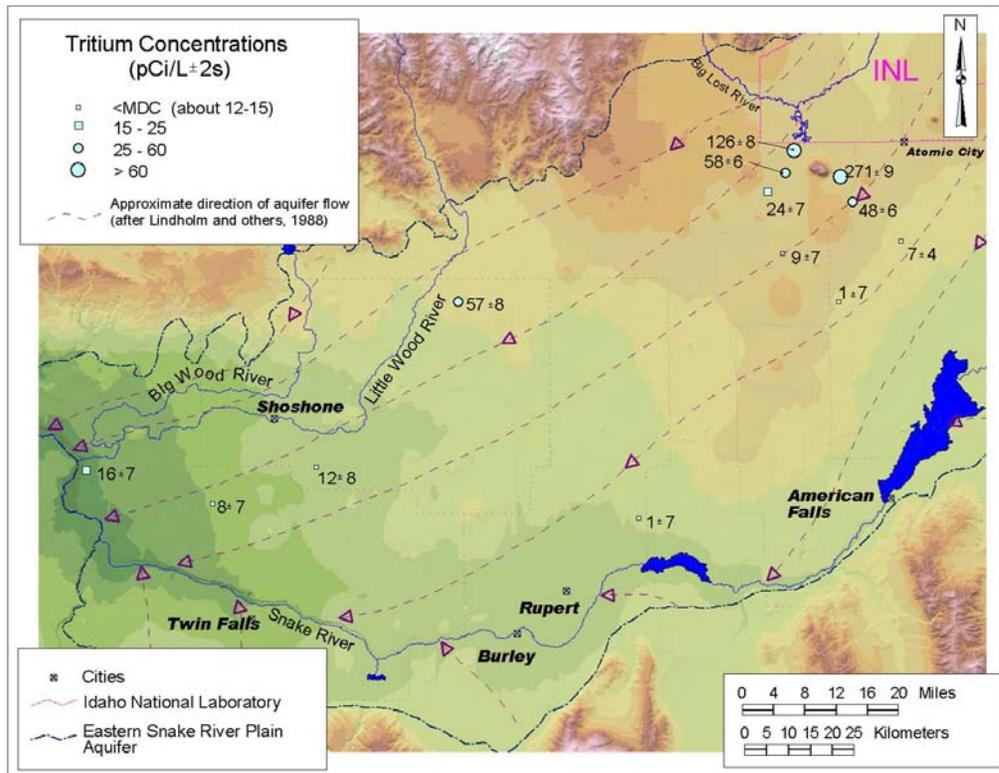


Figure 5. Tritium concentrations

Spatial plots of iodine-129 and tritium confirm that the highest concentrations of these known INL contaminants occur at the same sites. These sites are near (within a few kilometers) to the INL southern boundary. These plots also show that levels of these radionuclides vary for the locations sampled, even those sites determined to be outside of the influence of historic waste disposal at the INL. Thus, “background” for these radionuclides varies across the region sampled. The sites that likely have INL contaminants present include USGS 124, 125, 11, 14, and possibly the Houghland well. Other sites that may have concentrations of iodine-129 and tritium near the level observed in the sample site at the furthest edge of INL impacts, (the Houghland well) have other explanations for these radionuclides being present.

When considering all the results collected, many of the most abundant natural constituents in ground water (common ions and nutrients), most trace elements examined, and radiological measurements (gross-alpha and gross-beta radioactivity) are also components of wastes disposed



of historically by the INL. Even specific INL-identified waste products, such as iodine-129 or chlorine-36, can have a natural source and be concentrated by a natural process, such as evaporation of infiltrating water. Thus, the presence of these constituents, at slightly above-ambient concentrations, by themselves are insufficient to confirm INL impacts in the far-field. The spatial distribution of certain key indicators and the water chemistry context are both necessary to address the questions of where to look for INL impacts, how to identify those impact when they are seen, and how to separate those impacts from problems possibly due to well construction or local impacts.

Trends in Iodine-129

While direct disposal to the aquifer of many contaminants like iodine-129 has ceased, the impact of this disposal continues. Mann and Beasley (1994a) postulated that iodine-129 disposed of via an injection well at INTEC traveled the 26 km to USGS 14 in the 35 years from 1953 when disposal began until 1987 when it was extrapolated that iodine-129 concentrations exceeded their estimate of background. To further define the distribution of iodine-129 across the Eastern Snake River Plain Aquifer and refine estimates of “background”, Mann and Beasley collected additional samples during 1993-94. These are the data on which Cecil and others (2003) based their findings. The eight sites sampled in 1993 were primarily livestock wells in the region south of the INL while the remaining 24 sites, sampled in 1994, were from springs and surface water surrounding the Eastern Snake River Plain.

Six of the thirteen ground water sites sampled by the ESP during 1997-1998 correspond to sites sampled in 1993. Results for these sites, Coffee Point well, Houghland well, Crossroads well, Well #2, and USGS 11 and 14, were compared to assess changes in iodine-129 concentrations over time in this region of the Eastern Snake River Plain Aquifer. Results for a 1991 sampling effort were also available for two of these six sites.

Samples collected in 1991-1993 and reported by Cecil and others, (2003) were analyzed for iodine-129 using the AMS methods by IsoTrace Laboratory, at the University of Toronto, Toronto, Ontario, Canada. Samples were prepared at the Department of Energy Environmental Measurements Laboratory prior to shipping samples to IsoTrace for final preparation and analysis. Sample preparation and analysis are consistent with and comparable to analysis for iodine-129 at the PRIME Lab.

Results for 1991-1993 samples for the six sites compared ranged from $1.5 \pm 0.13 \times 10^{-6}$ to $42 \pm 1.7 \times 10^{-6}$ pCi/L, with 1991 results available for wells USGS 11 and 14. Sampling results for 1993 included data for USGS 14. Results for ESP sampling of these sites in 1998 ranged $0.16 \pm 0.013 \times 10^{-6}$ to $19 \pm 1.2 \times 10^{-6}$ pCi/L. (**Table 8**)



Method of Comparison

Statistical comparison of iodine-129 concentrations for DEQ-INL sampling in 1998 and selected sites sampled in 1991-1993 were made using a modification of the t-test for unequal variances. The t-test is described in Helsel and Hirsch (1992), pg 126. The modification follows that described in Cecil and others (1998).

The original equation for computing the test statistic, t where the sample variances differ is as follows:

$$t = \frac{\bar{X} - \bar{Y}}{\sqrt{\frac{S_x^2}{n} + \frac{S_y^2}{m}}} \quad (1)$$

Where:

- \bar{X} = the mean of the first group
- \bar{Y} = the mean of the second group
- S_x^2 = the variance of the first group
- S_y^2 = the variance of the second group
- n = the number of samples in the first group
- m = the number of samples in the second group
- t = the test statistic

This form of the t-test and its modification addresses the question “are the means of two sets of analyses the same?” (i.e. the null hypothesis $H_0: \mu_x = \mu_y$). The t-test assumes that the two sets of data are independent and normally distributed about their respective means. Additionally, the variances of the sets may differ. Analyses based on counting statistics, such as radioactive-decay counting techniques and mass spectroscopy methods such as AMS, where the ratio of a specific target isotope to stable isotope(s) are measured, are based on instrument counts over time. The reported analytical result is based on the mean and variance of a population of count rates. Where a sufficiently large number of counts were observed (<100), the distribution of counts, at least for radioactive decay counting, approximates a normal distribution (Currie, 1968).

The modification of Cecil and others (1998) is to set n and $m = 1$, replacing the mean of the samples (\bar{X}, \bar{Y}) and variance of the samples (S_x, S_y) with the analytical result and associated sample counting uncertainties, and assessing the equation for the absolute difference between X and Y where m and n are equal to 1 and a given test statistic t . The logic in setting m and n equal to 1 is based on the fact that the results X and Y are treated as one value with an associated counting uncertainty. The following equation is used for the comparison where m and n equal are equal to 1:



$$|X - Y| > t \left((S_x^2) + (S_y^2) \right)^{1/2} \quad (2)$$

Where:

X = analysis result for the first sample

Y = analysis result for the second sample

S_x = the counting uncertainty for the first sample

S_y = the counting uncertainty for the second sample

t = the critical test statistic for the chosen α -value, 1.96 is the test-statistic for the 95% confidence limit (Taylor, 1987, table C2)

Where the absolute value of the difference between X and Y exceeds the calculated value on the right-hand side of the equation, for the test-static of 1.96, the samples X and Y are determined to be different at the 95 percent confidence-level. If a test-static of 3.29 were used, the associated confidence level would be 99 percent. Assumed for this relationship is that the analytical result reported is the true mean and the counting uncertainty the true variance for the population of counts, and that the population of counts is distributed normally.

Comparison of Iodine-129 Results

Table 8 and **Figure 6** summarize the comparison of 1998 ESP AMS results for iodine-129 results with USGS 1991-1993 results. Based on a 95 percent confidence, results from all the sites were different. However, samples from the Houghland well did not differ at the 99 percent confidence level. The greatest difference between results was noted for samples collected from the Coffee Point well. ESP results were about 2 orders of magnitude lower for this site. For the ESP samples, the median was 3.0×10^{-6} pCi/L. The median for these USGS sites was 7.3×10^{-6} pCi/L. Thus, for the group of sites sampled, overall iodine-129 concentrations showed a decrease from 1991-93 to 1998.

Iodine-129 results for five of the six sites decreased from 1991-1993. An increase was observed for the Houghland well, from $4.5 \pm 0.22 \times 10^{-6}$ to $5.2 \pm 0.19 \times 10^{-6}$ pCi/L. Concentrations for wells USGS 11 and 14, reported as having INL iodine-129 contamination in Mann and Beasley (1994a, 1994b), decreased over the period between 1991-1993 and 1998. Samples were collected from USGS 14 for iodine 129 analysis in both 1991 and 1993, with results showing an increase from 1991 ($30 \pm 2.0 \times 10^{-6}$ pCi/L) to 1993 ($42 \pm 1.0 \times 10^{-6}$ pCi/L) before decreasing to $19 \pm 1.0 \times 10^{-6}$ pCi/L for 1998. Concentrations for USGS 11 decreased from 1991 ($10 \pm 1.0 \times 10^{-6}$ pCi/L) to $6.1 \pm 0.18 \times 10^{-6}$ pCi/L for 1998. A sample result for iodine-129 was not available for 1993 from USGS 11.



Table 8. Comparison of 1997-1998 ESP AMS iodine 129 analysis results from selected sites sampled by Mann and Beasley, 1990-1991 and 1993. Concentrations are expressed in pCi/L.

Well ID	ESP AMS			Mann and Beasley			Are results the same at 95% confidence
	Sample Date	(x10 ⁻⁶)		Sample Date	(x10 ⁻⁶)		
		I-129	± 1SD		I-129	± 1SD	
Coffee Point	6/18/98	0.16	0.013	6/21/93	10.0	0.39	No
Crossroads	6/23/98	0.76	0.026	6/22/93	3.2	0.19	No
Houghland	6/18/98	5.2	0.19	6/22/93	4.5	0.22	No
USGS 11	10/7/98	6.1	0.18	8/21/91	10.0	1.0	No
USGS 14	10/7/98	19.0	1.2	8/11/93	42.0	1.0	No
Well #2	6/18/98	0.20	0.012	6/21/93	1.5	0.13	No

The trend in concentrations for USGS 11 and 14 might possibly represent the passing of the 1958 waste disposal peak for iodine-129. Cecil and others (1998) indicated that the anticipated peak in chlorine-36 aquifer concentrations had reached well USGS 11, about 24 km south of the INTEC injection well, in about 1984, and USGS 14, about 26 km south of the INTEC injection well, in about 1987. Beasley and others (1998) examined the relative mobility of isotopes disposed of to the aquifer and determined that relative to chlorine-36, iodine-129 was retarded (i.e. did not travel in the aquifer as quickly). Cecil, Weldhan, and others (2000) estimated a travel time of about 2.5 to 3 meters per day for the 1958 disposal peak for chlorine-36 to reach USGS 11 and 14. Assuming that the iodine-129 disposal peak reached USGS 14 in about 1993 requires a travel time of 2 meters per day, much the same as the 1.8 meters per day suggested by Mann and Beasley (1994b). At 2 meters per day, approximately 50 years would be required for INL iodine-129 to travel the 40 km from the INTEC injection well to the Houghland well, and at 3 meters per day, about 36 years, thus, it is possible that iodine-129 concentrations at Houghland well are reflective of INL waste disposal. A similar argument can be made to explain the difference observed for the Crossroads well. This well is located about 30 km distant from the INTEC injection well, and only 6 km further than USGS 11. At an approximate travel time of 2 meters per day it is possible that the 1958 disposal peak had passed Crossroads well by 1998.

Iodine-129 concentrations for Coffee Point well and Well #2 also declined from 1993-1998. The largest relative differences were for Well #2 and Coffee Point well, differ by one to two orders of magnitude from the 1993 sampling. Review of chemistry and radiological results for these two sites offers some support to the assertion that general ground water flow is to the south and west from sites USGS 124 and USGS 14 towards the Houghland well. Well #2 and Coffee Point well would be perpendicular to this flow path. The strongest support is provided by the observed distribution of tritium in **Figure 5**, and chlorine-36 in **Figure 4**. The estimated ground water



contours published by Linholm and others (1988), and the simulated potentiometric contours published by Ackerman (1995) support the assumption that ground water flow is not from the direction of INL and INTEC and thus INL contaminants would not travel toward the wells in question. However, both predicted water level contours are relatively flat in that region and are not definitive. It is possible that the observed differences could be due to sample preparation and analysis-related factors that Mann and Beasley (1994a) discussed. These are errors in the sample preparation process, cross contamination in the sample analysis process, or natural variations that occur in the analysis process. Of these factors, the first two have the greatest potential to result in the differences observed for these samples. Such errors can not be accounted for or quantified with AMS measurements for iodine-129.

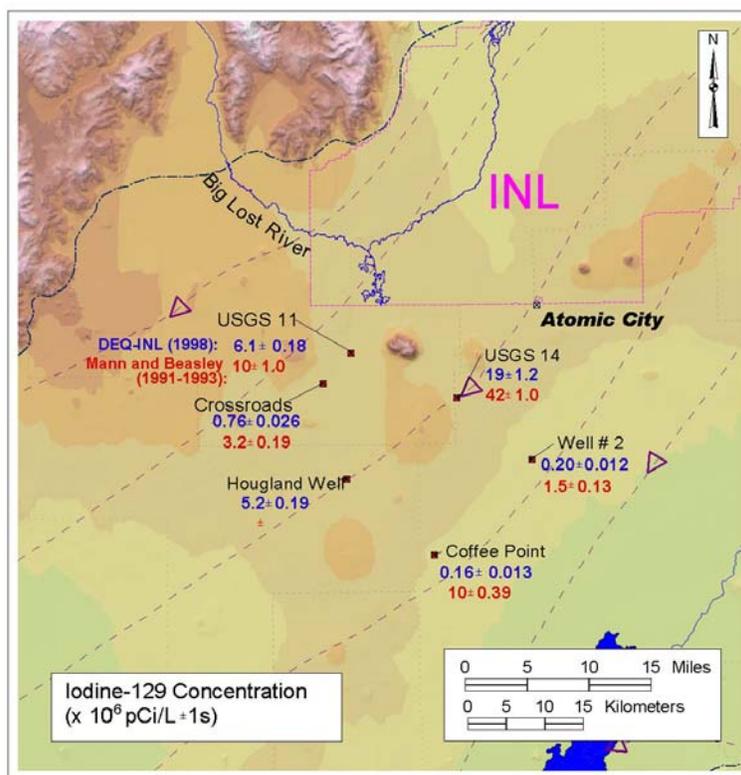


Figure 6. Comparison of results

Summary of Conclusions

Thirteen sites were sampled by the DEQ-INL during 1997-98. Results for selected trace metals, common ions, nutrients and radiological analytes were typically within expected ranges. Analysis for dissolved iodide showed that concentrations are somewhat higher than assumed by Mann and Beasley (1994b), with a median value of 0.005 mg/L compared to the assumption that concentrations are typically at or less than 0.001 mg/L for sites on the Eastern Snake River Plain Aquifer. None of the constituents sampled for approached any drinking water MCL.



Distribution of results for iodine-129, and tritium reflected two sources for these radionuclides--cosmogenic sources related to influence by infiltrating surface waters from the Wood River drainage, as evidenced by a combination of tritium and total nitrate concentrations; and historical INL waste disposal, evidenced by the distribution of tritium and iodine-129 concentrations, increasing towards the INL. Two wells interpreted to have INL iodine-129 and tritium also yielded chlorine-36 concentrations indicative of INL impact.

An adaptation of the two-sample t-tests for samples with different variances was used to compare iodine-129 results from samples collected during 1991-93 and samples collected in 1998 at the six comparable sites. At the 95 percent confidence level, samples from all sites differed. For all but one, the 1998 concentration was less. The median for these sites decreased from 7.3×10^{-6} pCi/L in 1991-93, to 3.0×10^{-6} pCi/L in 1998 samples. The greatest differences were noted between samples collected from Coffee Point well and Well #2. DEQ-INL results were one to two orders of magnitude lower for these sites.

Review of all information suggests that flow in the aquifer is to the south and west from the INL boundary. Flow is more towards the Houghland and Crossroads wells instead of the Coffee Point well and Well #2. Thus monitoring should be focused more in this south westerly direction.

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