EML-567

# Environmental Measurements Laboratory

# INVENTORY OF SITE-DERIVED $^{36}$Cl IN THE SNAKE RIVER PLAIN AQUIFIER, IDAHO NATIONAL ENGINEERING LABORATORY, IDAHO

T. M. Beasley
February 1995

DEPARTMENT OF ENERGY
NEW YORK, N. Y. 10014

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# ABSTRACT

Radioactive waste management practices at the U.S. Department of Energy's Idaho National Engineering Laboratory (INEL) in Idaho have introduced $^{36}$Cl ($T_{1/2}$ = 301,000 yr) into the Snake River Plain aquifer underlying the site. The $^{36}$Cl is believed to originate from neutron activation of stable $^{35}$Cl in nuclear fuels (principally) and in reactor cooling/process water.

Wastewater releases of $^{3}$H at the INEL have been documented by the site operators for the period 1952 to 1988. During this time, approximately 1.2 PBq of $^{3}$H (30,000 Ci) were introduced to the subsurface through disposal wells and seepage ponds. By sampling a number of monitoring and production wells downgradient from points of introduction, $^{3}$H movement and dispersion in the groundwater have been documented by the U.S. Geological Survey.

The present report uses these historical $^{3}$H release and monitoring data to choose hydrologic parameters (matrix porosity and plume penetration depth) that produce concordance between the $^{3}$H release estimates and the inventory calculated from measurements of $^{3}$H in the subsurface. These parameters are then applied to $^{36}$Cl isopleths to generate an estimated $^{36}$Cl inventory in the subsurface.

Using assumptions about irradiation times, neutron fluxes, and total fuel processed, as little as 23 g of stable chloride impurity in fuel elements would be adequate to produce the amount of $^{36}$Cl estimated to be in the groundwaters underlying the site.

The highest atom concentration of $^{36}$Cl measured onsite ($222 \times 10^{10}$ atoms $l^{-1}$) corresponds to an activity level of $\sim 4$ pCi $l^{-1}$ and represents 0.2 percent of the U.S. Environmental Protection Agency's (EPA) drinking water standard for this radionuclide (2000 pCi $l^{-1}$).

# ACKNOWLEDGMENTS

All of the water samples discussed here were supplied by the U.S. Geological Survey Project Office at the Idaho National Engineering Laboratory (INEL). L.D. Cecil and L.J. Mann warrant particular recognition in this respect.

The $^{36}$Cl measurements were performed at both the Nuclear Structure Research Laboratory of the University of Rochester and PRIME LAB, Purdue University. Drs. D. Elmore, P.W. Kubik, P. Sharma, U. Fehn and H. Gove deserve special thanks for their efforts in performing and overseeing these analyses during the week-long, 24-hour days that routinely attend the operation of these facilities during accelerator mass spectrometry runs.

The report has benefited from discussions and comments during preparation with L.J. Mann, B. Orr, D. Ackerman, and M. Luzinski (U.S. Geological Survey Project Office - INEL) who were always willing to share their understanding of hydrology and operational practices at the INEL with the author.

Lastly, the work reported here was supported by the Office of Environmental Restoration (DOE) at the Idaho Field Office thru a cooperative project with the U.S. Geological Survey Project Office (ERDI - 062-92) and by the Office of Health and Environmental Research (DOE).

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# INTRODUCTION

Between 1952 and 1984, low-level radioactive waste was discharged to the Snake River Plain aquifer at the Department of Energy's Idaho National Engineering Laboratory (INEL) by disposal wells and seepage ponds at two major facilities at the site. These were the Test Reactor Area (TRA; seepage ponds, principally) and the Idaho Chemical Processing Plant (ICPP; disposal well and seepage ponds). Figure 1 shows the location of these facilities, as well as others, and the spatial relationship between the facilities and the site boundaries. The ICPP operates as a nuclear fuel/waste water reprocessing facility; the TRA is the site for several experimental test reactors.

Beginning in 1961, and continuing to the present, the U.S. Geological Survey has monitored the presence and movement of selected radionuclides in the Snake River Plain aquifer which were introduced into the subsurface at the TRA and ICPP. The most comprehensive data base has been compiled for $^{3}$H; measurements of other activation and fission product radionuclides, while routinely performed, have been less extensively reported (Pittman et al., 1988). Mann and Cecil (1990) have recently summarized the groundwater $^{3}$H data at the INEL for the period 1961 through 1988, including release estimates for this radionuclide at both the TRA and the ICPP.

In 1987, $^{36}$Cl measurements of water from the U.S. Department of Energy's Savannah River Site (SRS) near Aiken, SC gave evidence of releases of this radionuclide from the fuel reprocessing facilities there (Beasley et al., 1992). In early 1990, exploratory $^{36}$Cl measurements were made in groundwaters and surface waters on and near the INEL to determine if $^{36}$Cl releases had occurred there as well. Excess $^{36}$Cl that has been detected in groundwaters downgradient from both the TRA and the ICPP can be ascribed to radioactive wastewater releases at these facilities (Beasley et al., 1992). Moreover, as at SRS, gaseous releases of $^{36}$Cl have also occurred at the ICPP (Cecil et al., 1992).

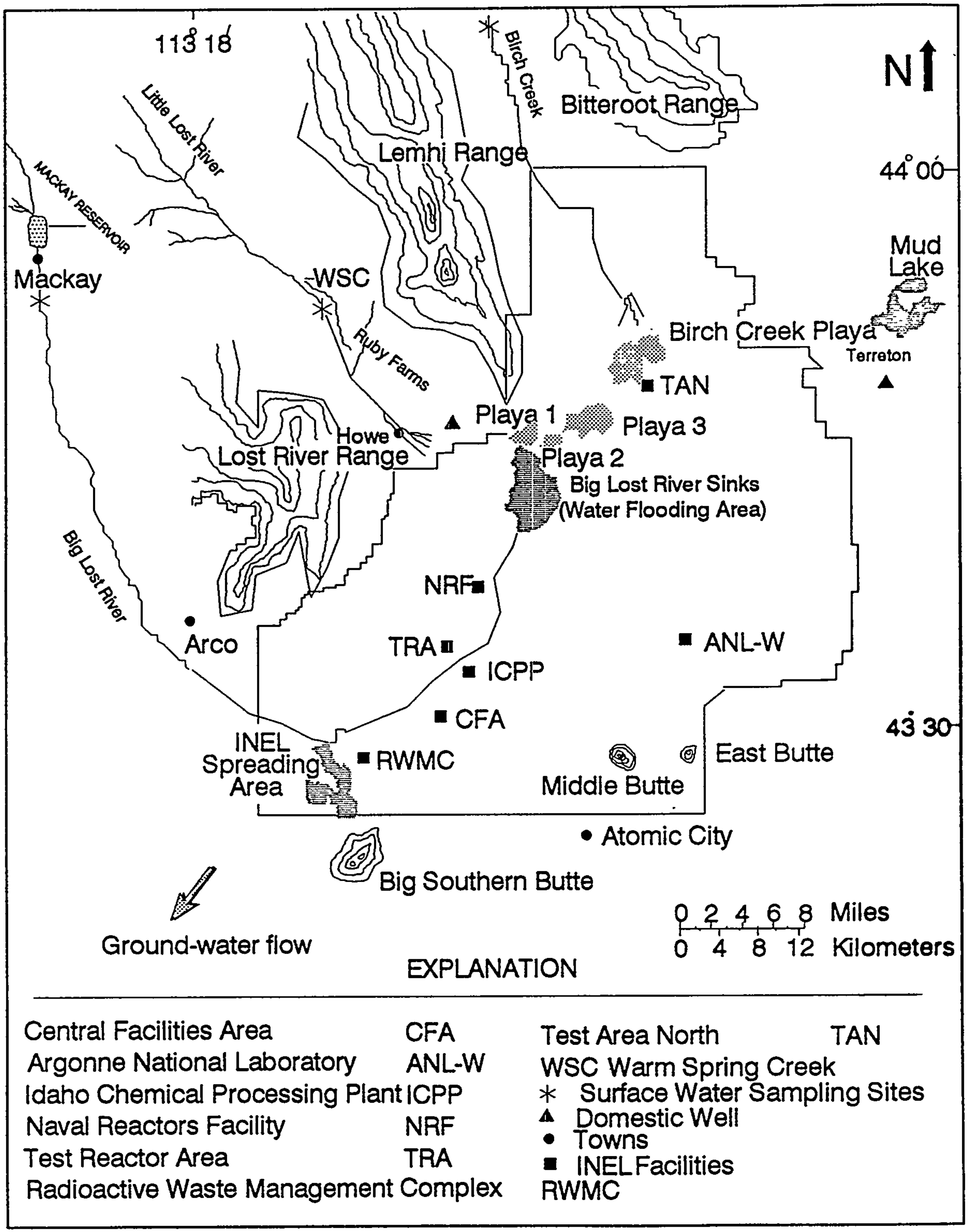

Figure 1. Selected Facilities and Surface Water Sampling Sites
Idaho National Engineering Laboratory (adapted from Mann and Cecil, 1990).

A suite of groundwater samples has been analyzed for $^{36}$Cl in an attempt to demonstrate the utility of using this radionuclide as a groundwater tracer at the site and to construct an inventory of $^{36}$Cl releases to the ground water comparable to the present-day data base that exists for $^{3}$H. However, unlike $^{3}$H, there were no $^{36}$Cl measurements made at the INEL in any medium prior to 1990. Therefore, in order to approximate a $^{36}$Cl inventory, hydrogeologic parameters which give concordance between the inventory of $^{3}$H in the subsurface, as estimated from historical release data, and a calculated inventory based on the areal distribution of $^{3}$H in the site groundwaters have been used. These hydrogeologic parameters include: 1) the effective porosity of the vesicular basalts which comprise the bulk of the geological matrix of the Snake River Plain aquifer at the INEL, and; 2) the depth to which the contaminated water spreads and penetrates into the saturated zone over the length of its flow path. Where concordance is demonstrated between the historical releases and calculated subsurface inventories of $^{3}$H, it is assumed that the same hydrogeologic parameters can be used to construct an inventory for $^{36}$Cl, because both radionuclides behave conservatively in the subsurface (Bentley et al., 1986).

## BACKGROUND

The INEL, formerly called the National Reactor Testing Station (NRTS), began operation in 1949. Over its history, a number of prototype reactors (52) have been constructed, tested, and operated there for military, industrial and research purposes. In addition, the ICPP is a multipurpose facility used primarily for recovering highly enriched uranium from a variety of fuels used in naval propulsion, research, and test reactors (Benedict et al., 1981). Between 1953 and 1984, low-level radioactive wastewater was discharged from the ICPP directly to the Snake River Plain aquifer by means of a 183-m deep disposal well. In early 1984 this practice was discontinued and wastewater is now discharged to seepage ponds where the water infiltrates through the vadose zone to the aquifer approximately 140 m below the land surface. At the TRA, radioactive wastewater has historically been discharged to seepage ponds. At both facilities, perched water zones have been identified which contain radio-labeled water (Pittman et al., 1988).

The Snake River Plain aquifer is unconfined and flows through a complex sequence of basalt flows of Quaternary age; the tops of the flows are highly fractured and hydraulic conductivities are correspondingly high. The upper 150 m of these formations are thought to be the most permeable to flow (Lindholm, 1988). Ground-water trajectory across the site is from the northeast to the southwest (Figure 1) at flow rates estimated to vary between 1.5 to 6.0 m day$^{-1}$ (Robertson et al., 1974).

Between 1952 and 1988, approximately 1.2 PBq (30,000 Ci) of $^3$H were released in wastewater from the TRA and the ICPP, of which some 0.38 PBq were estimated to remain in the Snake River Plain aquifer by 1988 (Mann and Cecil, 1990). Because there are no well clusters that sample sequential horizons in the Snake River Plain aquifer downgradient from either the TRA or the ICPP, it is not possible to experimentally apportion the $^3$H inventory in three dimensions within the saturated zone. However, the $^3$H inventory has been presumed to reside in the upper 60 m of the basalt flows because wells open to the aquifer below this depth do not yield samples with $^3$H concentrations above the reporting level of the analysis (0.5 pCi ml$^{-1}$; Mann and Cecil, 1990). Moreover, there is evidence that vertical hydraulic heads in the aquifer increase slightly with depth leading to possible net upward flow of water (Mann, 1992).

Figure 2 shows existing wells completed in the Snake River Plain aquifer as of 1988; Figures 3 through 7 (adapted from Mann and Cecil, 1990) show the lateral and longitudinal development of the tritium plume at the INEL between 1961 and 1988. In 1961 both the tritium detection limit and the number of wells available for sampling limited the delineation of the plume to the dimensions and $^3$H activities shown. With advances in radiation detection instrumentation and additional well construction,

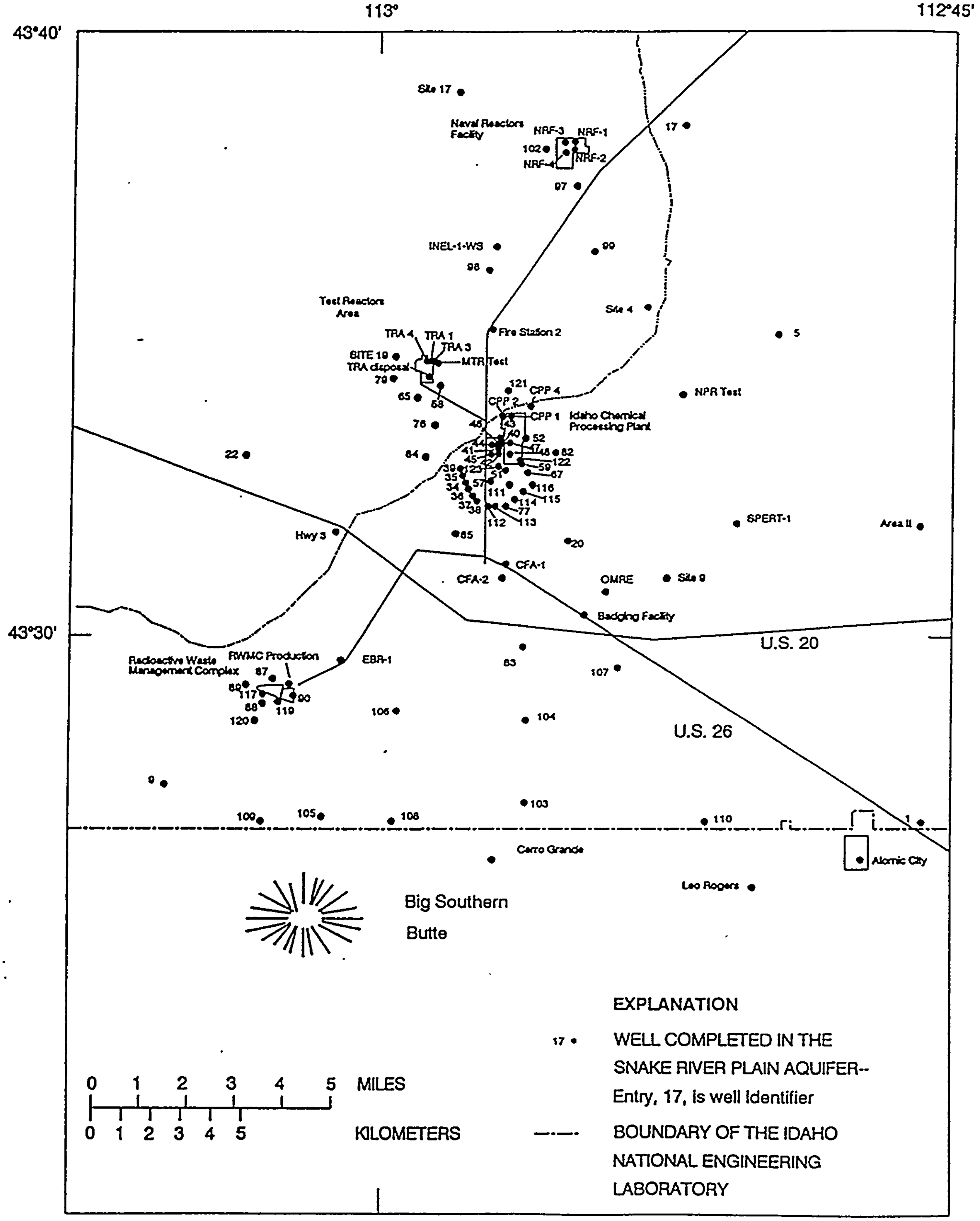

Figure 2. Monitoring and Production Wells Completed in the Snake River Plain Aquifer (from Orr and Cecil, 1991)

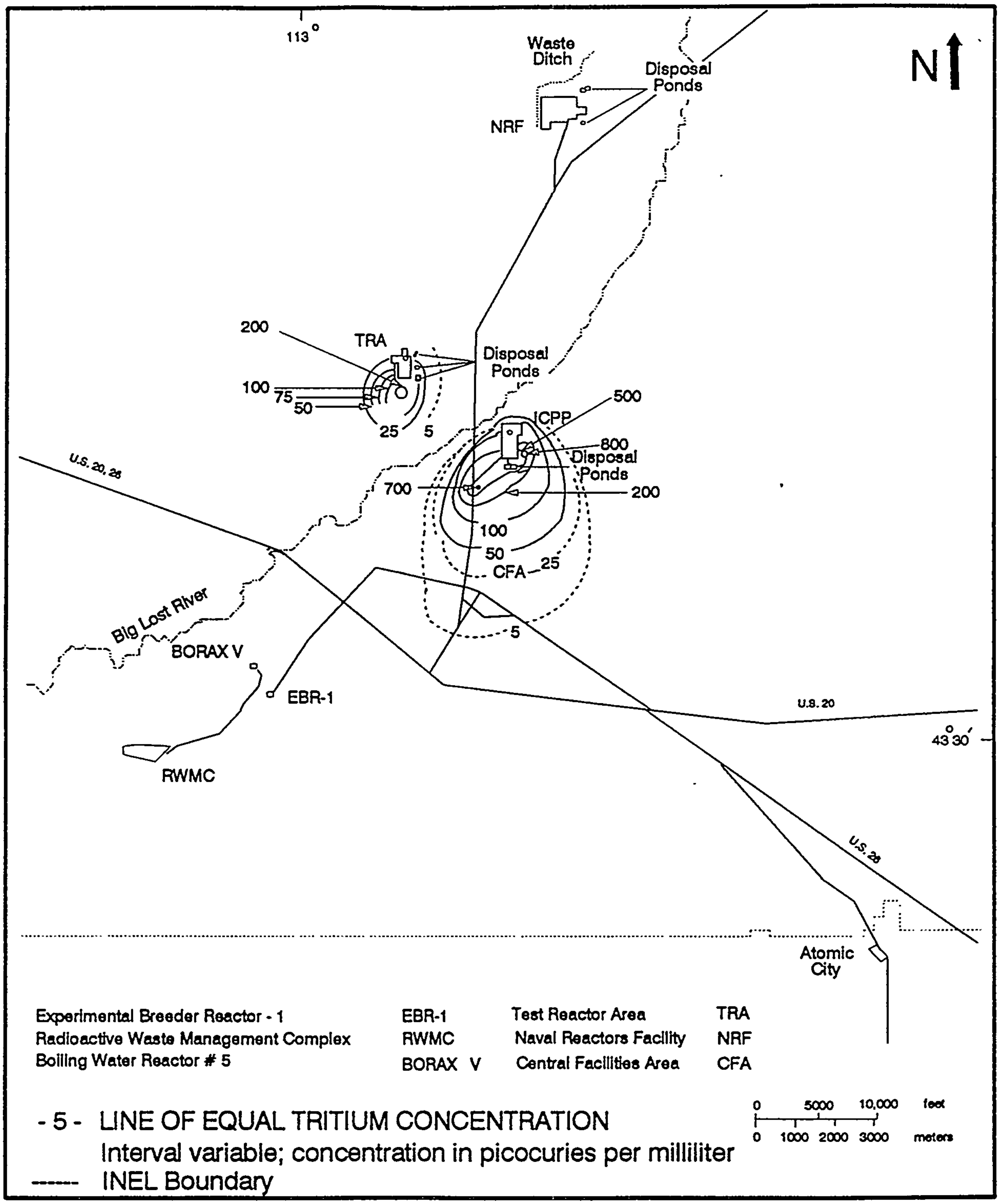

Figure 3. Tritium Isopleths - Snake River Plain Aquifer (1961).
(adapted from Mann and Cecil, 1990).

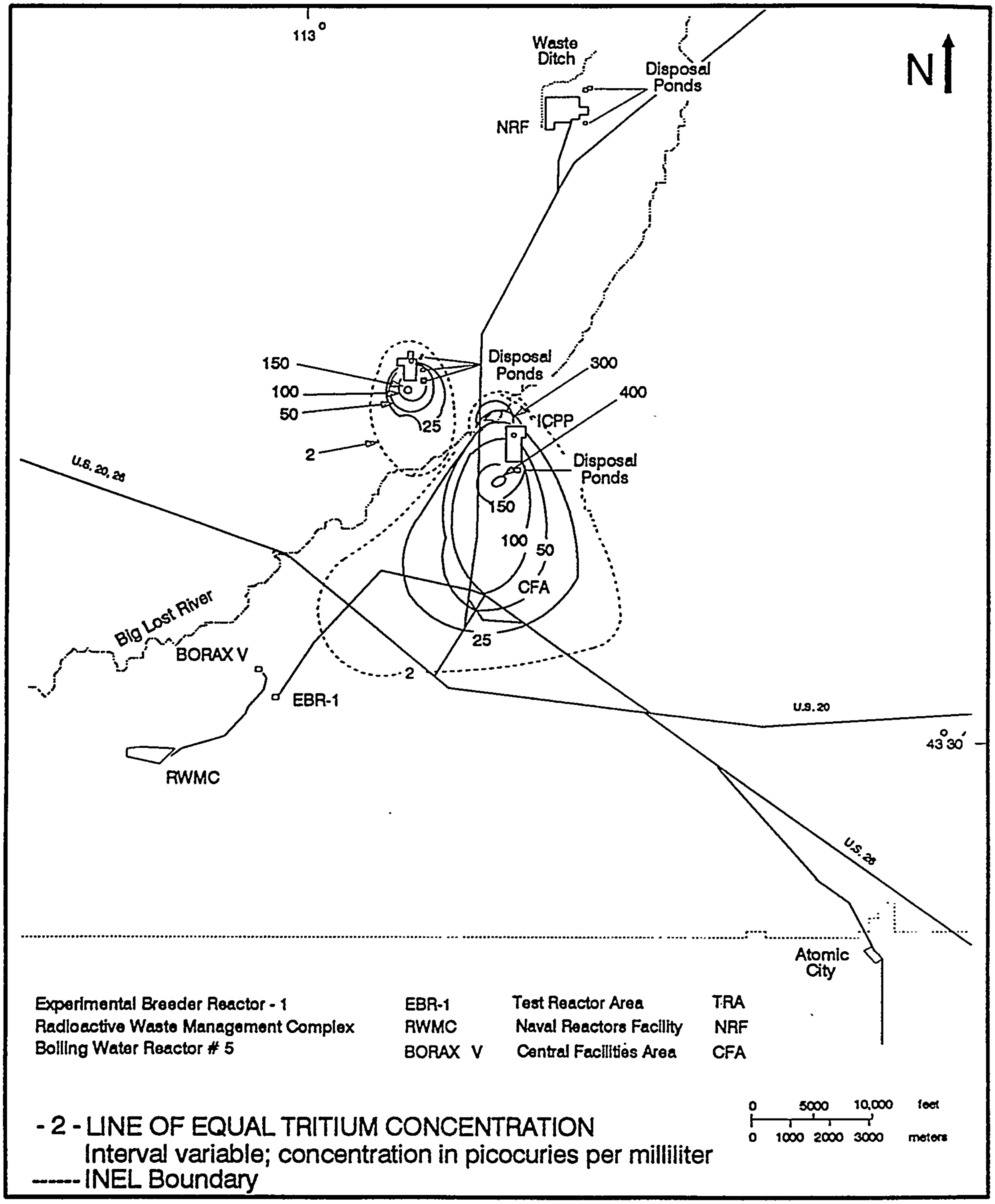

Figure 4. Tritium Isopleths - Snake River Plain Aquifer (1970).
(adapted from Mann and Cecil, 1990).

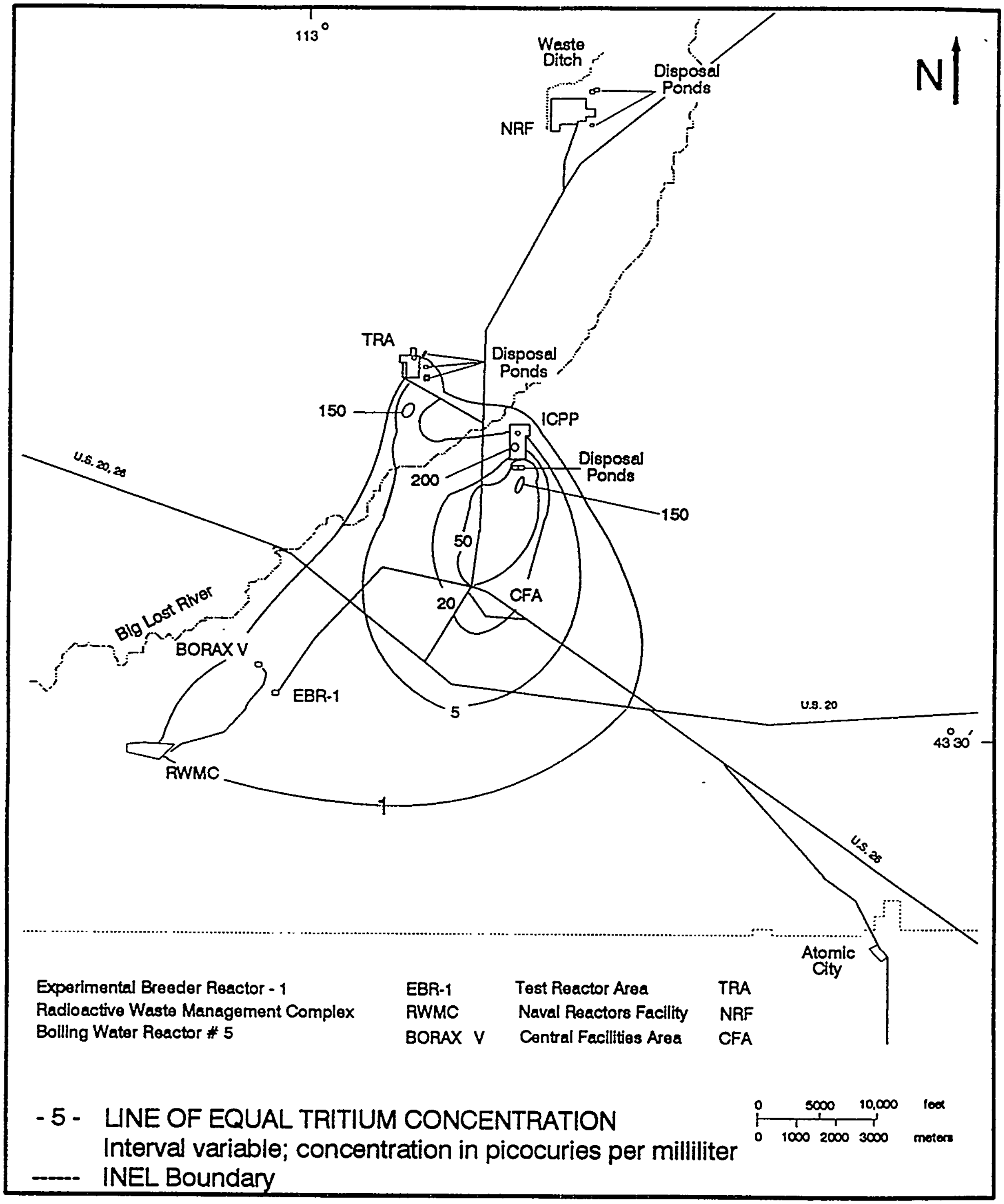

Figure 5. Tritium Isopleths - Snake River Plain Aquifer (1977).
(adapted from Mann and Cecil, 1990).

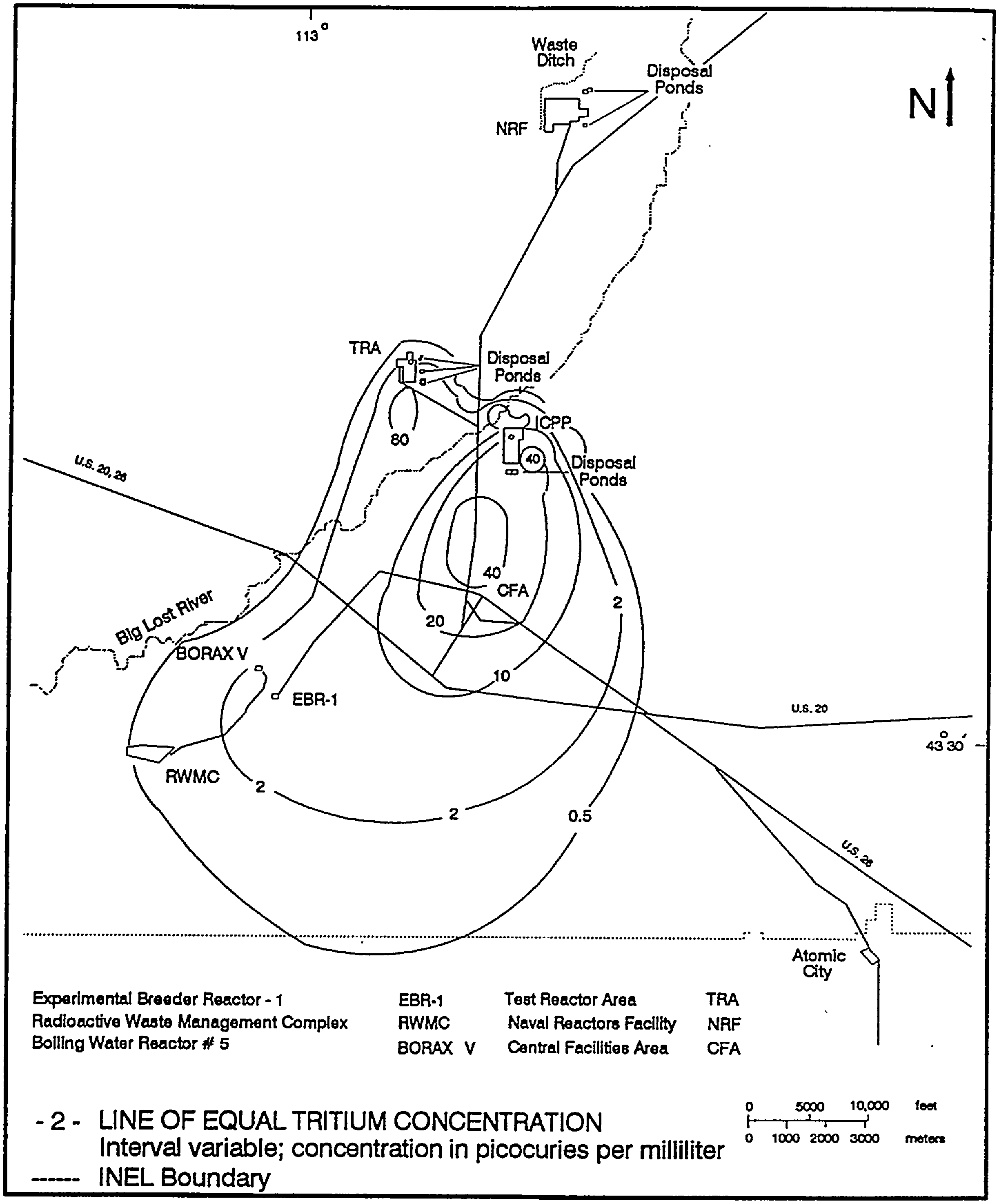

Figure 6. Tritium Isopleths - Snake River Plain Aquifer (1985). (adapted from Mann and Cecil, 1990).

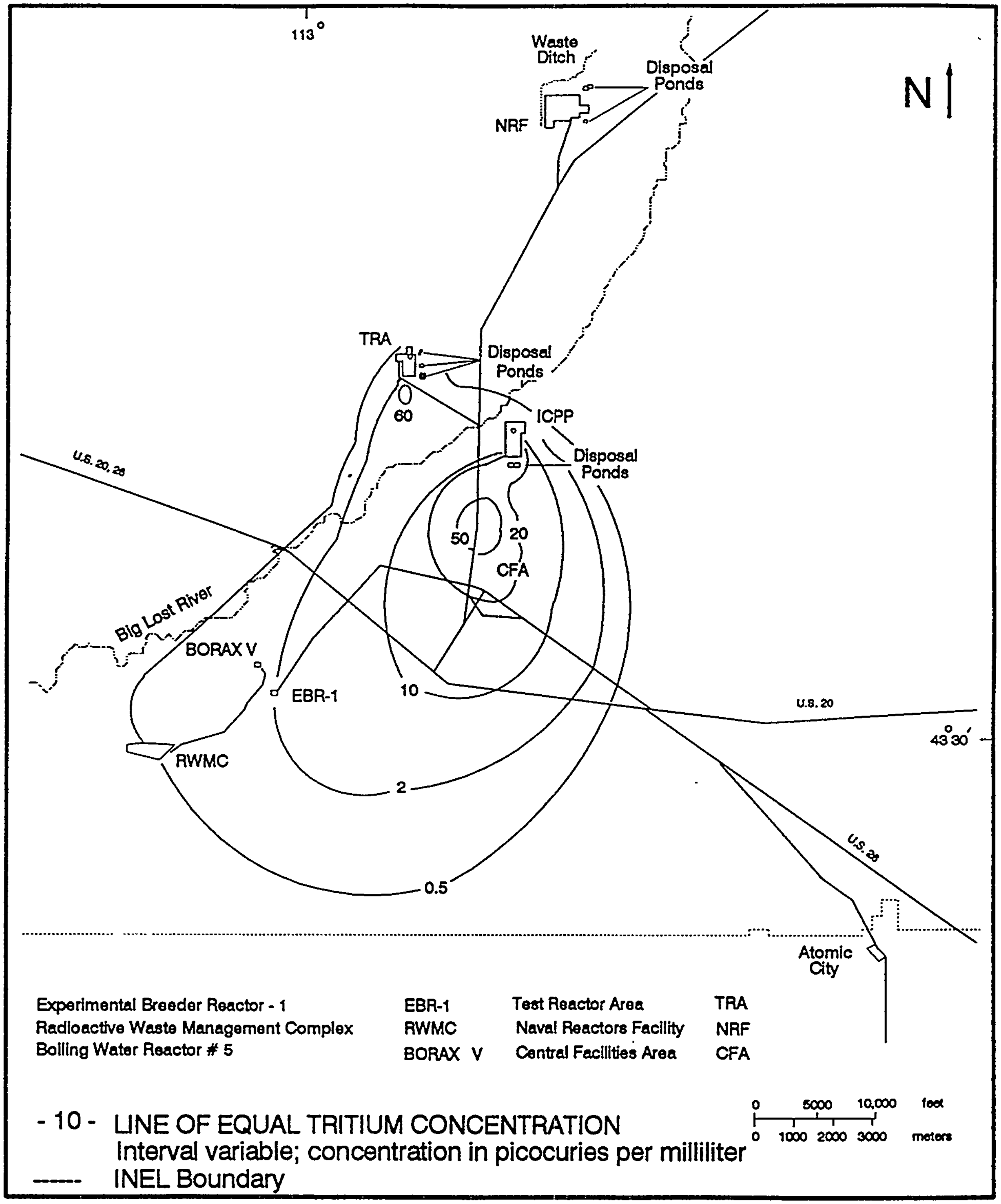

Figure 7. Tritium Isopleths - Snake River Plain Aquifer (1988).
(adapted from Mann and Cecil, 1990).

refinements in the plume definition improved to the degree shown by the 1985-1988 $^3$H isopleths.

The shape of the $^3$H plume at the INEL has been the subject of several modeling papers which use the dimensional development of the $^3$H isopleths over time to assign numerical values to hydrodynamic parameters of the flow system. Once estimated, these parameters can then be used to predict the continued development of the plume into the next century.

The transport of a conservative solute through saturated, fractured media and the change in solute concentration with distance from the release point are functions of the effective porosity of the media, (interconnected pore spaces), the flow velocity, and the coefficient of hydrodynamic dispersion (Bear and Verruijt, 1987). For one-dimensional flow, the coefficient of hydrodynamic dispersion is given by the expression:

$$D_l = \alpha_l \bar{v}_X + D^* \qquad (1)$$

where:

$D_l$ = longitudinal coefficient of dispersion (length$^2$/time)
$\alpha_l$ = longitudinal dispersivity (length)
$\bar{v}_X$ = the average linear ground-water velocity (length/time)
$D^*$ = molecular diffusion

Equation (1) shows that the longitudinal dispersion of the solute is a function of both mechanical dispersion and molecular diffusion, processes that cannot be distinguished in groundwater that is in motion. For a two dimensional system, where the flow velocity varies along the flowline and the coefficients of dispersion vary through space, the change in the solute concentration with time is given as (Freeze and Cherry, 1979):

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x_l}\left(D_l \frac{\partial C}{\partial x_l}\right) + \frac{\partial}{\partial y_t}\left(D_t \frac{\partial C}{\partial y_t}\right) - \frac{\partial}{\partial x_l}\left(\bar{v}_x C\right) \qquad (2)$$

where:

$C$ = solute concentration (arbitrary units);
$\bar{v}_x$ = mean linear velocity along flow line in x direction (m $yr^{-1}$); and,
$D_{l,t}$ = longitudinal and transverse coefficients of dispersion ($m^2$ $yr^{-1}$)

It is generally observed in both laboratory and field studies that $\alpha_l$ is typically greater than $\alpha_t$ (transverse dispersivity) for solute plumes that develop in flowing groundwaters in porous media. In those cases where $\alpha_l \gg \alpha_t$, spreading of the plume longitudinally is pronounced and the plume is elongated and oblong; the shape of the INEL $^3H$ plume has consistently suggested that $\alpha_t$ and $\alpha_l$ are of similar magnitude (Robertson, 1974; Lewis and Goldstein, 1982; Duffy and Harrison, 1987; Goode and Konikow, 1990). Robertson (1974) was able to approximate the early shape of the $^3H$ plume using a steady-state, two dimensional numerical model in which: 1] transmissivity over the flow path was assumed constant; 2] storativity was constant and uniform in space, and; 3] $\alpha_t$ and $\alpha_l$ were assigned values of 140 and 90 m, respectively. Robertson attributed the transposition in the relative magnitude of the dispersivities to spatial heterogeneity within the fractured basalt leading to pronounced horizontal, transverse dispersive mixing. Goode and Konikow (1990) argued that transients in the flow field occasioned by periods of unusual recharge in the Big Lost River can lead to imprecise estimates in both $\alpha_t$ and $\alpha_l$. They recalibrated Robertson's model (using additional data collected after 1972) to include the effects of hydraulic head changes resulting from Big Lost River recharge events. Interestingly, the ability to predict chloride concentrations downgradient from the disposal well at the ICPP (Figure 1) using either steady-state or transient flow conditions proved to be relatively insensitive to dispersivities over a wide range of values.

The importance of the dispersivities in predicting solute spreading, both horizontally and vertically, can be illustrated by reference to the modeling of solute behavior in porous media by Pickens and Lennox (1976).

They used equation (2) to simulate the two-dimensional movement of a conservative solute introduced into a steady-state, saturated groundwater system. Figure 8A shows the distribution of the tracer following 15 years of flow where the hydraulic conductivity was 0.5 m day$^{-1}$, the porosity was set equal to 0.3, and the longitudinal and transverse dispersivities ($\alpha_{l,t}$) were varied. It is significant to note that where the ratio of dispersivities remains constant, but increases in magnitude, the tracer can move deep into the flow system. Figure 8B illustrates the effect of increasing the magnitude of the transverse dispersivity while holding the horizontal dispersivity constant. Where $\alpha_t \geq \alpha_l$, the theoretical depth of penetration can be even more pronounced (Pickens and Lennox, 1976). It must be reemphasized, however, that in modeling the solute distributions shown in Figure 8, vertical flow resulting from differences in the vertical hydraulic head between the water table and depth within the saturated zone is assumed to be negligible, i.e., $y_z = 0$.

The calculation of radionuclide inventories within a plume requires that the depth of radionuclide penetration with distance can be approximated. From the foregoing discussions, it is clear that this cannot be done unambiguously, especially for the basalts underlying the INEL. There are no well clusters downgradient from either the TRA or the ICPP that draw water from different depth horizons thereby permitting estimates of the vertical penetration of radioactivity into the aquifer. In the past, a maximum, 60-meter penetration depth has been assumed for the $^3$H plume (Mann and Cecil, 1990) based on undetectable $^3$H in waters raised from wells exceeding this depth (Wells 83 and EBR-1; Figure 1). However, the $^{36}$Cl/Cl ratio in one well, EBR-1 (Appendix 2) is significantly higher than those of other onsite wells [outside the influence of the $^3$H plume] for comparable Cl$^-$ ion concentrations (Appendix 3), suggesting an additional source of $^{36}$Cl to this well. The well bore for EBR-1 penetrates approximately 150 m of the aquifer; well 83 exhibits only "background" $^{36}$Cl/Cl ratios even though it lies directly within the $^3$H and $^{36}$Cl plumes and

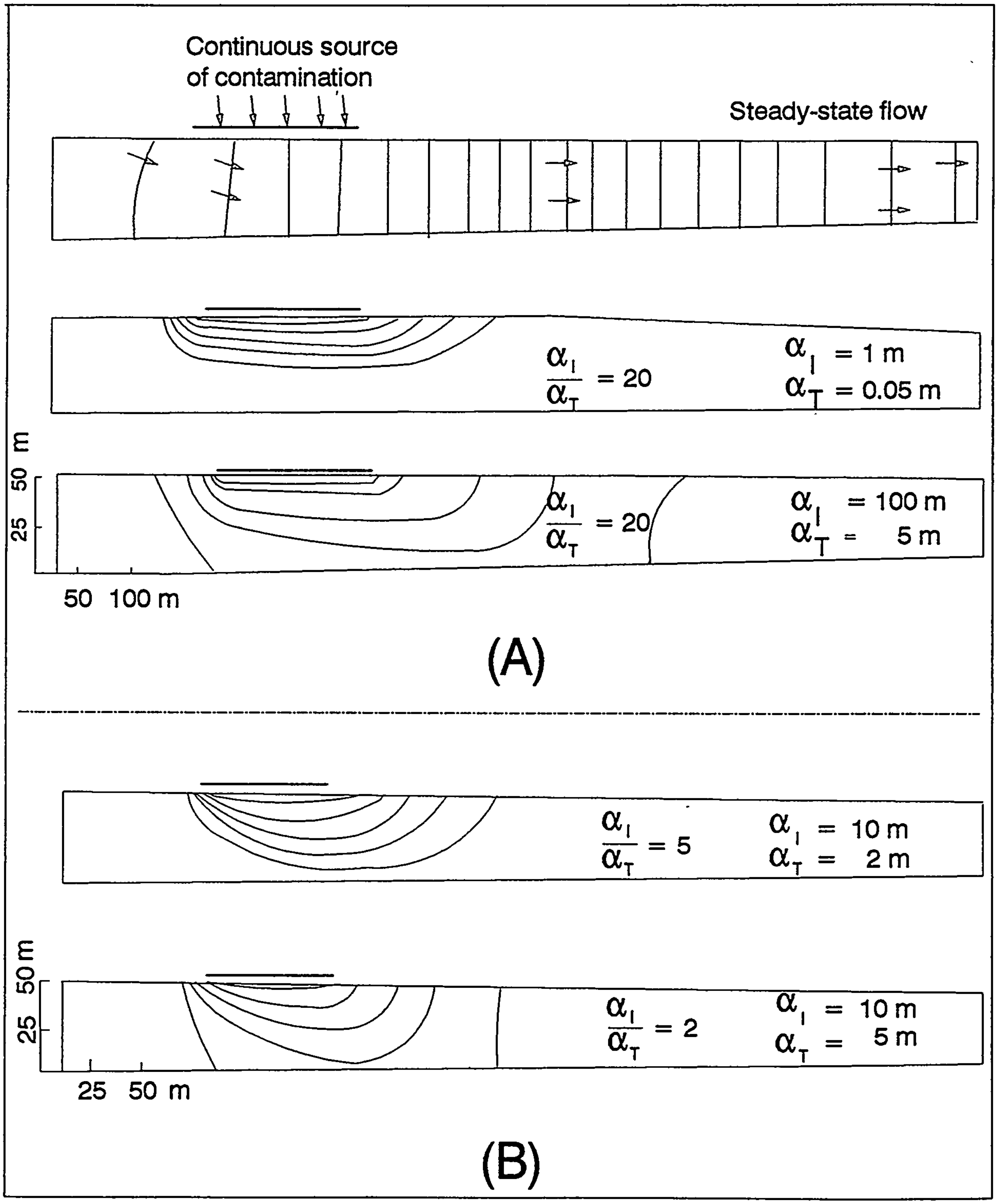

Figure 8. Concentration Distribution as a Function of Longitudinal and Transverse Dispersivity. Contours Represent $C/C_O$ of 0.9, 0.7, 0.5, 0.3, and 0.1, Respectively Simulation of Conservative tracer; t = 15 yr (adapted from Pickens and Lennox, (1976)

draws water from depth intervals comparable to that for well 104 ( $\sim$ 72 m) where $^{36}Cl/Cl$ ratios are substantially elevated (Appendix 2). These data serve to reinforce the complexities that surround the estimation of subsurface flow in highly fractured media.

Regardless of the uncertainties in the plume penetration depths that are selected for inventory purposes, if primary reference is made to the historical releases of $^{3}H$ from the TRA and the ICPP, then choosing penetration depths that yield an integrated inventory comparable to that released will produce a $^{36}Cl$ inventory whose uncertainty is comparable to the uncertainties that attend the $^{3}H$ release estimates.

## METHODS

The chemical isolation and purification techniques that were used to prepare the targets and the AMS measurement protocols have been described elsewhere and are not repeated here (Beasley et al., 1992).

Table 1 shows the cumulative, decay corrected $^{3}H$ releases over time at the TRA and the ICPP (Mann and Cecil, 1990). It should be noted that in cases where $^{3}H$ in waste streams was released to seepage ponds, no estimate of evaporative loss of the radionuclide has been attempted. Since the half-life of $^{3}H$ is 12.3 years (Lederer and Shirley, 1978), approximately 5.5 percent of the $^{3}H$ decays annually. By 1988, of the cumulative 30,858 Ci of $^{3}H$ estimated to have entered the Snake River Plain aquifer, some 10,200 Ci remained.

To illustrate the calculational methods, the 1988 isopleths of $^{3}H$ in the Snake River Plain aquifer (Figure 7) have been assigned penetration depths between 50 and 60 meters. Recent measurements of $^{129}I$ from packer tests at well 47 have confirmed that the majority of the $^{129}I$ is contained in waters within this depth interval (Mann, 1993). To simplify the calculation of

# TABLE 1

ANNUAL RELEASES OF $^3H$ FROM THE TEST REACTOR AREA (TRA) AND THE IDAHO CHEMICAL PROCESSING PLANT (ICPP) ACTIVITIES IN CURIES
(from Mann and Cecil, 1990)

| Year | ICPP | TRA | Total | $\sum$ |
|---|---|---|---|---|
| 1952 | -- | 190 | 190 | 190 |
| 1953 | 456 | 190 | 646 | 836 |
| 1954 | 608 | 190 | 798 | 1,634 |
| 1955 | 808 | 190 | 998 | 2,632 |
| 1956 | 1,543 | 190 | 1,733 | 4,365 |
| 1957 | 969 | 190 | 1,159 | 5,524 |
| 1958 | 3,504 | 190 | 3,694 | 9,218 |
| 1959 | 2,565 | 190 | 2,755 | 11,973 |
| 1960 | 679 | 190 | 869 | 12,842 |
| 1961 | 590 | 190 | 780 | 13,622 |
| 1962 | 361 | 279 | 640 | 14,262 |
| 1963 | 1,084 | 353 | 1,437 | 15,699 |
| 1964 | 1,768 | 412 | 2,180 | 17,879 |
| 1965 | 97 | 328 | 425 | 18,304 |
| 1966 | 250 | 398 | 648 | 18,952 |
| 1967 | 861 | 424 | 1,285 | 20,237 |
| 1968 | 510 | 522 | 1,032 | 21,269 |
| 1969 | 125 | 749 | 874 | 22,143 |
| 1970 | 75 | 653 | 728 | 22,871 |
| 1971 | 59 | 342 | 401 | 23,272 |
| 1972 | 298 | 176 | 474 | 23,746 |
| 1973 | 32 | 184 | 216 | 23,962 |
| 1974 | 455 | 240 | 695 | 24,657 |
| 1975 | 43 | 260 | 303 | 24,960 |
| 1976 | 43 | 293 | 336 | 25,296 |
| 1977 | 734 | 140 | 874 | 26,170 |
| 1978 | 316 | 126 | 442 | 26,612 |
| 1979 | 225 | 105 | 330 | 26,942 |
| 1980 | 109 | 135 | 244 | 27,186 |
| 1981 | 359 | 191 | 550 | 27,736 |
| 1982 | 209 | 515 | 724 | 28,460 |
| 1983 | 436 | 203 | 639 | 29,099 |
| 1984 | 12 | 161 | 173 | 29,272 |
| 1985 | 393 | 261 | 654 | 29,926 |
| 1986 | 251 | 77 | 328 | 30,254 |
| 1987 | 215 | 128 | 343 | 30,597 |
| 1988 | 89 | 172 | 261 | 30,858 |
| Total | ~21,100 | ~9,700 | | |

contaminated volumes, the areas assigned to each individual isopleth have been assigned single, fixed penetration depths. The effective porosity of the basalt is assigned a value near 0.23 based on recent laboratory measurements of Bishop (1991) who measured both effective and total porosity of a large block of basalt quarried near the Radioactive Waste Management Complex (RWMC; Figure 1). Therefore, for any given area, the $^3$H inventory was calculated as the product of the area bounded by the isopleths, the assumed penetration depth, the effective porosity and the $^3$H activity within the isopleths. Because gradients exist in the $^3$H activities with distance, individual areas were subdivided and assigned $^3$H activities that were calculated as the average activity between successive isopleths. As an example, for the 1988 $^3$H inventory (Figure 7), the area bounded by the 20 and 10 pCi ml$^{-1}$ isopleths was divided into two equal areas; the dividing line was assumed to have a $^3$H activity concentration of 15 pCi ml$^{-1}$ and therefore the average $^3$H activity concentrations in the two subdivided areas were assigned values of 17.5 and 12.5 pCi ml$^{-1}$, respectively.

By 1988, the $^3$H inventory in the groundwater downgradient from the TRA and the ICPP, from releases at those facilities, is calculated to be ~ 10,200 Ci. Table 2 sets out the variables that were used to estimate $^3$H activities contained within the plumes shown in Figure 7 and the resultant, summed inventory. The calculated inventory, ~ 10,240 Ci agrees very well with the expected inventory of 10,200 Ci. Because the operational measurement limit for $^3$H is 0.5 pCi ml$^{-1}$, the integrated inventory does not include areas where activities are lower than this value. However, including an area of 50 x 10$^6$ m$^2$ (equal to that of the 0.5 pCi ml$^{-1}$ isopleth; Table 2) with penetration depths of 60 meters and concentrations of 0.25 pCi ml$^{-1}$ increases the $^3$H inventory by only 173 Ci. Accepting the uncertainties discussed earlier, the penetration depths chosen produce concordance between the calculated and expected inventories.

# TABLE 2

## INTEGRATION VARIABLES FOR $^{3}H$ INVENTORY ESTIMATES - 1988

| Isopleth | Area ($10^6$ $m^2$) | Depth (m) | Mean Activity Concentration (pCi $ml^{-1}$) | Total Activity (Ci) |
|---|---|---|---|---|
| 60 | 0.2 | 50 | 60 | 138 |
| 50 | 1.5 | 50 | 50 | 1050 |
| | 1.9 | 50 | 40 | 874[§] |
| 20 | 1.9 | 50 | 30 | 655 |
| | 1.9 | 50 | 20 | 437 |
| 10 | 8.2 | 60 | 17.5 | 1815[§] |
| | 8.2 | 60 | 12.5 | 1297 |
| 2 | 23 | 60 | 7.5 | 2182[§] |
| | 23 | 60 | 3.5 | 1018 |
| 0.5 | 25 | 60 | 1.5 | 517[§] |
| | 25 | 60 | 0.75 | 258 |
| TOTAL | 120[†] | | | 10,241 |
| EXPECTED | | | | 10,200 |

---

[†] $120 \times 10^6$ $m^2$ = 46 $mi^2$

[§] The 20, 10, 2, and 0.5 pCi $ml^{-1}$ isopleths were divided into the respective areas shown and assigned average activities as explained in the text (see Figure 7 for isopleth areas)

The choice of 50-meter penetration depths for radioactive wastewater introduced to the aquifer at the ICPP disposal well requires some justification because, as constructed, the casing was perforated at two depth intervals (Mann and Beasley, 1993). One interval ranged from 126 to 138 m while the other interval was from 149 to 181 m. Depth to water at this location is approximately 138 m. Thus, while wastewater was introduced above the water table, comparable to water originating from infiltration ponds at the TRA, water was also introduced at depth. However, the operational history of the ICPP disposal well has not been without difficulty. On three separate occasions, between 1953 and 1982, casing failures and well bore shallowing by sediment loading required well rehabilitation. Because of these difficulties, the exact depth zones over which wastewater was introduced into the aquifer are unknown. It is likely, however, that given the frequency of these occurrences, the bulk of the wastewater was injected at depths between 126 m and 152 to 168 m (Mann and Beasley, 1993).

## RESULTS AND DISCUSSION

Appendix 2 lists the *in-situ* $^{36}$Cl/Cl ratios, $^{36}$Cl atom concentrations and stable chloride concentrations for the onsite wells that were sampled to estimate the subsurface $^{36}$Cl inventory resulting from waste disposal practices at the TRA and the ICPP. Appendix 3 contains results for onsite wells uninfluenced by releases from either the TRA or the ICPP while Appendix 7 lists results for perched water in the vicinity of the TRA.

Figure 9 shows the resultant, onsite isopleths of $^{36}$Cl constructed from the data of Appendix 2. In many aspects, the shapes of the individual isopleths show a strong correspondence to those for $^{3}$H in 1988. It is clearly apparent, however, that using $^{36}$Cl as a tracer of contaminated water movement across the INEL southern boundary can extend the distance to which downgradient plume migration can be followed. Indeed, two wells (Wells 11 and 14, Appendix 2, Figure 11) which lie approximately 6 and 12 km due south of the INEL boundary have $^{36}$Cl/Cl ratios and $^{36}$Cl atom concentrations that are above "background". The U.S. Geological Survey is

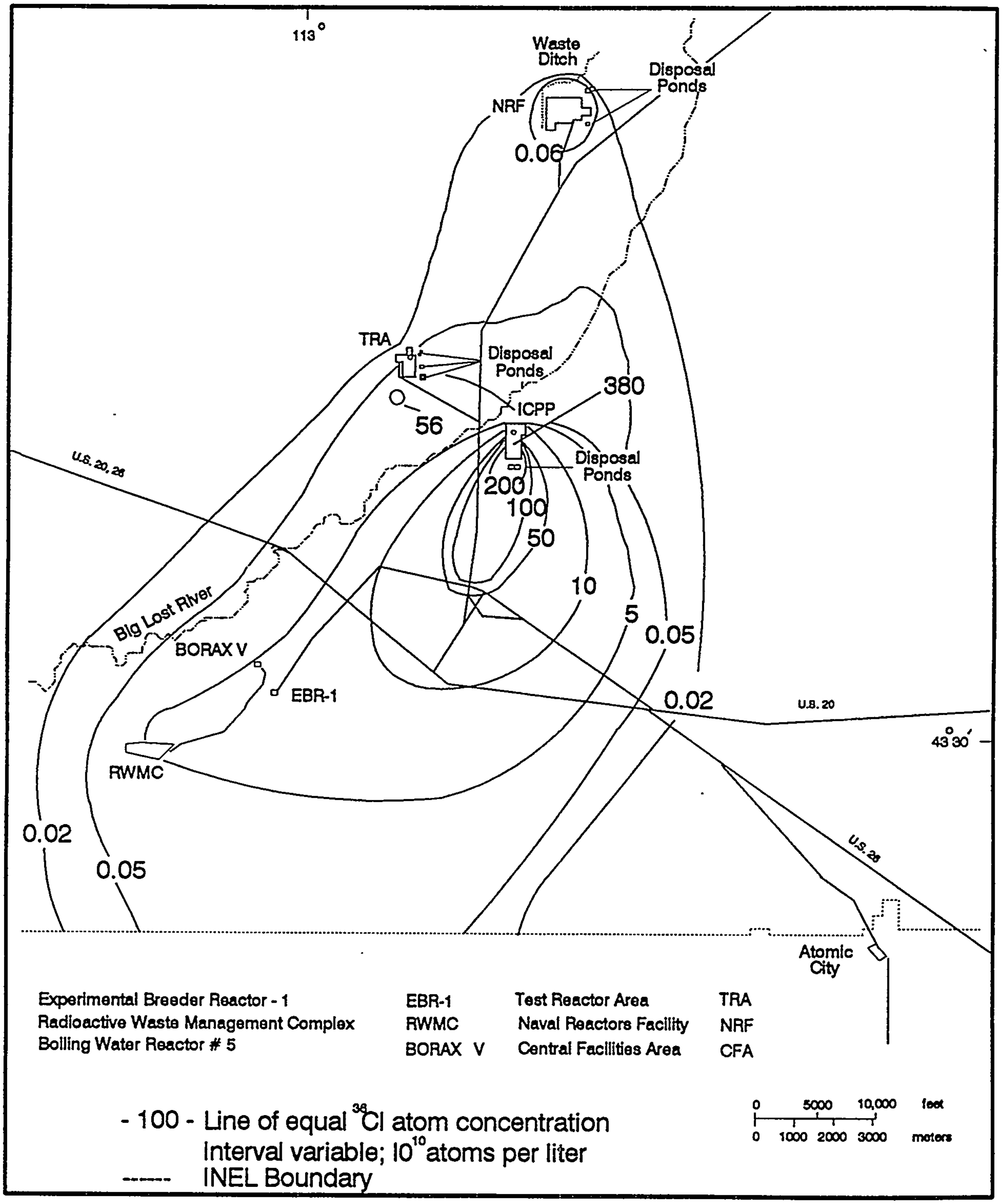

Figure 9. Chlorine-36 Isopleths - Snake River Plain Aquifer (1992).

presently drilling two new wells between the site boundary and Wells 11 and 14 (anticipated completion dates: 1994 and 1995) to corroborate that the $^{36}$Cl concentrations at Wells 11 and 14, provisionally attributed to past discharges at the TRA and the ICPP, represent extensions of the $^{36}$Cl plume shown in Figure 9.

For wells outside the $^{3}$H plume (Appendix 3, Figure 10), $^{36}$Cl/Cl ratios generally range between ( $\sim 300$ to $\sim 600$) x $10^{-15}$, values which are consistent with those of meteoric origin at the latitude of the INEL (Bentley et al. 1986) . The mean atom concentration of $^{36}$Cl in background water is near $10^{8}$ atoms $l^{-1}$. Surface water and selected wells away from the INEL (Appendix 4, Figure 1) tend, on the average, to have mean $^{36}$Cl atom concentrations that are about 40 percent higher than those of the uncontaminated, onsite wells although the standard deviations about these means make such comparisons tenuous. It is not unreasonable, however, to posit that residual bomb-pulse $^{36}$Cl in the drainage basins of the streams and rivers sampled is still being mobilized by interbed flow to these waters (Beasley et al., 1992). The Arco City Well derives recharge from the Big Lost River which would explain the similarity in their $^{36}$Cl/Cl ratios; the elevated $^{36}$Cl atom concentrations in the Ruby Farms Well and the much lower $^{36}$Cl atom concentrations in Birch Creek have been explained elsewhere (Beasley et al., 1992).

To the southwest of the INEL site, $^{36}$Cl/Cl ratios and $^{36}$Cl atom concentrations in wells of the eastern Snake River Plain are generally representative of meteorically-derived $^{36}$Cl (Appendix 5, Figure 11). There are, however, exceptions. It has already been noted that Wells 11 and 14 show evidence of site-derived $^{36}$Cl in those waters (Appendix 2). The same can be argued for both the Houghland and Crossroads Wells. At the distal end of the Plain, only wells MV-37 and MV-39 give indication of "excess" $^{36}$Cl as reflected in their $^{36}$Cl/Cl ratios. A reasonable explanation for this observation is that both wells are likely influenced by infiltration of waters from Big Wood River and Little Wood River which lie to the north and south of the wells (Mann, 1995). Waters from these rivers could be expected to carry bomb-$^{36}$Cl as runoff within their respective drainage basins.

There is strong evidence that small but measurable releases of $^{36}$Cl have occurred at the Naval Reactors Facility, most probably in water released to the waste ditch there (Figure 9). From 1982 to 1985, some 93 million gallons yr$^{-1}$ of wastewater were released to the ditch. Once released, the water infiltrates through the vadose zone to the aquifer below. Contained in this water were large amounts of chloride, sulfate, and sodium ion with average concentrations near 400, 100, and 100 ppm, respectively (Pittman et al., 1988). As with the TRA and the ICPP, the source of the $^{36}$Cl in this water can reasonably be attributed to the thermal neutron activation of stable $^{35}$Cl.

Using the same integration technique and variables as those previously described for $^{3}$H, the 1990/1991 onsite inventory of $^{36}$Cl downgradient from the TRA and the ICPP is calculated to be approximately 19 gigaBecquerel (GBq) or $\sim$ 0.5 Ci. The area occupied by the onsite plumes is approximately 240 x $10^{6}$ m$^{2}$ ( $\sim$ 92 mi$^{2}$). The radioactivity estimates include subtraction of meteorically-derived $^{36}$Cl which is important only for calculating the $^{36}$Cl inventory in the plume downgradient from the NRF. For example, assuming that the $^{36}$Cl/Cl ratio from natural, atmospheric production is 600 x $10^{-15}$ (range = 300 to 600 x $10^{-15}$; Bentley et al., 1986), and that the "background" concentration of chloride ion in groundwater at the INEL is approximately 15 ppm (range = 8 to 15 ppm; Robertson et al., 1974), the maximum concentration of naturally-derived $^{36}$Cl is calculated to be 1.5 x $10^{8}$ atoms l$^{-1}$. The plume downgradient from the TRA and the ICPP holds some 2.4 x $10^{23}$ atoms of $^{36}$Cl; of this total, a maximum of 6 x $10^{20}$ atoms are meteorically derived.

Even though the offsite, subsurface inventory of site-derived $^{36}$Cl remains to be estimated following additional well construction, it is most probably less that 5 percent of the onsite inventory. For example, assigning an area to the offsite plume equal to that onsite ( $\sim$ 195 x $10^{6}$ m$^{2}$), penetrating to a depth of as much as 150 m, and having a mean $^{36}$Cl atom concentration of 0.1 x $10^{10}$ atoms $l^{-1}$ would produce a calculated inventory of ~ 0.5 GBq ( ~ 0.014 Ci) or about 3 percent of the onsite inventory (of which 0.071 GBq [0.002 Ci] could be from meteorically-derived $^{36}$Cl).

There are data from the Test Area North (TAN, Figure 1) that indicate small, but measurable amounts of $^{36}$Cl have been introduced to the aquifer in that area of the site. However, assigning a source of the $^{36}$Cl to a specific operations is not possible since most of the activities there have been of a classified nature. Nevertheless, for completeness, Appendix 6 lists data from wells sampled there (Figure 12) which can be compared to results from wells down- gradient from TRA and ICPP.

While the great majority of the research described here has been directed to understanding site-derived $^{36}$Cl additions to the Snake River Plain aquifer, areas of contaminated, perched water exists at the INEL. One such area is near the TRA (Pittman et al., 1988) where contaminated water released to the disposal ponds are slowed in their infiltration through alluvium at an alluvium/basalt interface some 100 to 150 feet below the land surface. Water raised from wells that sample the perched zone contain elevated levels of $^{36}$Cl (Appendix 7, Figure 13), however, the estimated $^{36}$Cl inventory of these waters is not large. For example, using data from Pittman et al. (depth of perched zone, 45 m; area of perched zone, 1.5 km$^2$; and porosity of alluvium, 0.3) and assigning a mean $^{36}$Cl atom concentration to these waters of 1.3 x $10^{10}$ atoms $l^{-1}$ (Appendix 7), the $^{36}$Cl inventory is calculated to be 2.6 x $10^{20}$ atoms, i.e., approximately 0.1 percent of the estimated $^{36}$Cl inventory in the Snake River Plain aquifer downgradient from the TRA and the ICPP.

## $^{36}$Cl PRODUCTION

It is instructive to estimate the total amount of stable chloride needed to produce 19 GBq of $^{36}$Cl by neutron activation of stable chloride. To do so, it would be necessary to know the neutron fluxes (thermal and epithermal) and irradiation times to which the chloride had been subjected. As stated earlier, the ICPP has processed fuels from a variety of government-owned reactors (Benedict et al., 1987). This processing has occurred over a period of 36 years during which time reactor design changes have led to progressive improvements in power density and therefore fuel loadings so that the estimation of an average flux and irradiation time that would be representative for $^{36}$Cl production calculations is tenuous at best. It is possible, however, to broadly illustrate the amounts of stable chloride required to produce the amount of $^{36}$Cl seen in the subsurface at the INEL by reference to reasonable assumptions concerning fluxes, irradiation times, and total fuel processed.

The transformation of stable target atoms in a particle flux can be expressed as:

$$N_1 = N_0\, e^{-\phi\sigma t} \qquad (3$$

where:

$N_1$ = number of target atoms remaining after time t;
$N_0$ = number of original target atoms;
$\sigma$ = cross section for $N_0$, in barns (1 barn = $10^{-24}$ cm$^2$);
$\phi$ = particle flux (particles cm$^{-2}$ sec$^{-1}$); and,
t = irradiation time (sec)

In the case of $^{36}$Cl production by thermal neutron irradiation of $^{35}$Cl the cross section is 43 barns (Lederer and Shirley, 1978). For epithermal neutrons, ( $\sim$4 keV) the resonance cross section is slightly less than 9 barns and near 44 keV, the cross section drops to near 0.4 barn (Macklin, 1984).

If the new atoms produced, $N_2$ , have a measurable (n,$\gamma$) cross section, then the number of new atoms produced at the end of a given irradiation period can be calculated from the expression (West, 1958):

$$N_2 = \frac{\sigma_1 N_0}{\sigma_2 - \sigma_1} (e^{-\phi\sigma_1 t} - e^{-\phi\sigma_2 t}) \tag{4}$$

where:

$N_0$ = number of target atoms $N_0$ at $t=0$;
$N_2$ = number of new atoms produced;
$\sigma_2$ = n,$\gamma$ cross section of new atoms, $N_2$ ($cm^2$);
$\sigma_1$ = n,$\gamma$ cross section of target atoms, $N_0$ ($cm^2$);
t = irradiation time (sec); and,
$\phi$ = particle flux (particles $cm^{-2}$ $sec^{-1}$)

Equation (3) is used to predict target burn-up when target atoms disappear only as a result of particle capture. For the $^{35}Cl$ (n,$\gamma$) $^{36}Cl$ reaction, rewriting equation (3) as:

$$\frac{N_1}{N_0} = e^{-\phi\sigma t} \tag{5}$$

gives the fraction of the target atoms remaining after an irradiation time, t, in a constant neutron flux, $\phi$. Table 3 shows the fraction of the original atoms of $^{35}Cl$ remaining after different irradiation times for two different thermal neutron flux levels. Thus, one way to estimate the amount of stable chloride activated to produce the amount of $^{36}Cl$ estimated to be in the aquifer downgradient from the TRA and the ICPP ( $\sim 3 \times 10^{23}$ atoms) would be to assume that, on average, fuels processed at the ICPP have an irradiation time of 4 years in a thermal neutron flux of $10^{15}$ n $cm^{-2}$ $sec^{-1}$. Under these conditions, the $^{35}Cl$ atoms are virtually depleted (they can never be entirely depleted because of the logarithmic nature of their removal) and are therefore approximately equal to the number of $^{36}Cl$ atoms produced.

## TABLE 3

### FRACTION OF STABLE $^{35}$Cl ATOMS REMAINING FOLLOWING IRRADIATION WITH THERMAL NEUTRONS§

| Irradiation Time (yr) ($\phi = 10^{14}$ n cm$^{-2}$ sec$^{-1}$) | $\frac{N_1}{N_0}$ | Irradiation Time (yr) ($\phi = 10^{15}$ n cm$^{-2}$ sec$^{-1}$) | $\frac{N_1}{N_0}$ |
|---|---|---|---|
| 1 | 0.87 | 1 | 0.26 |
| 2 | 0.76 | 2 | 0.07 |
| 3 | 0.66 | 3 | 0.017 |
| 4 | 0.58 | 4 | 0.004 |

§ The cross section ($\sigma$) is assumed to be 43 x $10^{-24}$ cm$^2$.

This assumes that once produced, $^{36}$Cl atoms themselves are not transmuted in the flux. Thus, approximately 3 x $10^{23}$ atoms of $^{35}$Cl ( $\sim$23 g of chloride) would have been irradiated. If an assumed 30 metric tons of fuel were processed each year, on average, during the 36 year operating history of the ICPP, the initial minimum chloride ion concentration of the fuels required to supply this amount of chloride would be $\sim$0.02 ppm (parts per million; $\mu$g g$^{-1}$). Because the capture cross section for epithermal neutrons decreases from 43 to 9 barns, using the same irradiation time and particle fluences, but assuming epithermal rather than thermal neutrons, raises the required minimum chloride concentration to $\sim$0.03 ppm. Irradiations at particle fluxes of $10^{14}$ n cm$^2$ sec$^{-1}$ for four years with thermal and epithermal neutrons yield calculated, lower limit chloride concentrations in the fuel which range between 0.05 and 0.2 ppm, respectively.

The thermal neutron capture cross section for $^{36}$Cl is not well known. It has been listed variously as having values near 100 barns (Heath, 1970) to less than 1 barn (Lederer and Shirley, 1978); accurate values for both thermal and epithermal resonance capture cross sections are yet to be determined (Holden, 1992). However, because the lower cross section (< 1 barn) is believed to be more realistic than the higher value (Holden, 1992), calculations using equation (4) indicate that the resultant production of 3 x $10^{23}$ atoms of $^{36}$Cl would require a total $^{36}$Cl production some 11 percent greater in order to account for its transmutation in a thermal neutron flux of $10^{15}$ n $cm^{-2}$ $sec^{-1}$ with an irradiation time of 4 years. This in turn raises the required amount of irradiated chloride by the same percentage - an insignificant amount considering other uncertainties inherent in the calculations.

Additions of $^{36}$Cl to the groundwater at the INEL arising from: a) $^{36}$Cl-labeled process waters released to the seepage ponds at the TRA or; 2) to $^{36}$Cl/Cl- labeled wastewater from TRA operations that are processed at the ICPP would reduce the amount of chloride in fuel elements necessary to produce the $^{36}$Cl inventory discussed above. Unfortunately, even order-of-magnitude estimates of this input are not available in the literature. Reconstruction of the history of $^{36}$Cl releases at the TRA and the ICPP is anticipated, using curatorial water samples from 1961 forward, which should help place this contribution in perspective (Cecil, 1991).

Finally, as stated earlier, it is now known that $^{36}$Cl atmospheric releases have occurred at the ICPP (Cecil et al., 1992). Some fraction of these releases may result from processing wastewater from the TRA and other facilities on the site (Hernandez, 1992). The remaining fraction would place an additional requirement for increased stable chloride concentrations in fuel elements processed there to account for these atmospheric releases. At present, no quantitative assessments of the atmospheric $^{36}$Cl releases at the INEL have been attempted and, therefore, it is not possible to estimate the amount of irradiated stable chloride required to account for such releases.

## CONCLUSIONS

Radioactive waste management practices at the INEL have introduced approximately 19 GBq of $^{36}$Cl into the Snake River Plain aquifer over the period 1952 to 1990. This inventory is approximately 0.005 percent of the $^{3}$H inventory (0.38 PBq) estimated to reside in the aquifer as of 1988. Neutron irradiation of trace amounts of stable chloride in both reactor fuel elements and reactor cooling/process waters are believed to be the major sources of the $^{36}$Cl released at the site.

Although there are similarities in the shapes of the $^{3}$H and $^{36}$Cl "plumes", the areal extent of the discernible $^{36}$Cl plume is significantly larger than that of $^{3}$H owing to the greater analytical sensitivity of measuring $^{36}$Cl by accelerator mass spectrometry techniques. Consequently, with the completion of additional wells south of the site boundary, future $^{36}$Cl measurements will permit a re-estimation of the flow velocity of contaminated water away from the southern INEL site boundary. At present, these velocities have been calculated on the basis of "first appearance" of measurable $^{3}$H in wells downgradient of the TRA and the ICPP, assuming that $^{3}$H releases to the Snake River Plain aquifer began in 1961. On the basis of such measurements, calculated flow velocities range between 1.5 and 6.0 m day$^{-1}$ (Robertson et al., 1974). The data from $^{36}$Cl suggest that these estimates may increase by as much as a factor of 2 when averaged over the entire flow trajectory.

Packer tests in selected wells at the INEL are periodically conducted to experimentally determine the depth to which labeled water penetrates the Snake River Plain aquifer following release from the TRA and the ICPP. Measuring both $^{3}$H and $^{36}$Cl in these tests will permit refined estimates of both the longitudinal and transverse dispersivities associated with water flow in the fractured basalts at the INEL. The data could also be used to paramaterize three dimensional flow models for solute transport within the aquifer.

Lastly, none of the $^{36}$Cl atom concentrations measured to date pose any radiological hazard to people on or offsite at the INEL. For example, Well 51 (Appendix 2) had the highest $^{36}Cl$ atom concentration measured, i.e. 222 x $10^{10}$ atoms $l^{-1}$. This atom concentration corresponds to an activity of 0.16 Bq $l^{-1}$(4.4 pCi $l^{-1}$) and represents approximately 0.2 percent of the Environmental Protection Agency (EPA) drinking water standard for this radionuclide which is about 2000 pCi $l^{-1}$(DOE, 1990). For the offsite wells 11 and 14, the measured $^{36}Cl$ atom concentrations (Appendix 2) correspond to less than 0.00001% of the EPA drinking water standard.

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# APPENDIX 1

## QUALITY ASSURANCE/QUALITY CONTROL PROTOCOLS

The sensitivity of the AMS technique for measuring $^{36}$Cl precluded taking more than a few milliliters of raw water for analysis in many of the wells sampled so as not to contaminate the ion source at the AMS facilities where the analyses were performed. Because the stable chloride ion concentrations at these volumes was not sufficient to produce an adequate AgCl target for the analysis, 24 mg of Weeks Island Halite (as chloride) was added to the samples, before processing, to produce ample target material. Process blanks for all of analyses using 18 megohm-cm water and Weeks Island Halite carrier consistently yielded targets with $^{36}$Cl/Cl ratios $< 10 \times 10^{-15}$; in no series of samples analyzed was there any indication of cross contamination between samples.

Stable chloride concentrations were determined by two methods: 1] titration with mercuric chloride, and; 2] ion chromatography techniques. Standards for both methods were prepared, separately, by dissolving appropriate amounts of 99.9999% pure NaCl in 18 megohm-cm water. Agreement between methods, for the same sample, was generally within ± 5 percent; agreement between duplicates using a single technique was generally within ± 2 to 3 percent (Beasley et al, 1992).

Known and blind duplicates were periodically prepared and analyzed at the AMS facilities to confirm the reproducibility of the measurements. Table 4 shows the results of these analyses and demonstrates, over a two year period, the satisfactory agreement between duplicates run at both the Nuclear Structure Research Laboratory and PRIME LAB; in almost every instance, the agreement is within ± 15 percent.

# TABLE 4

# REPRODUCIBILITY OF AMS MEASUREMENTS FOR $^{36}$Cl/Cl IN KNOWN AND BLIND DUPLICATES

| Well/Blind I.D. | $^{36}$Cl/Cl (x $10^{-15}$) | |
|---|---|---|
| | NSRL | PRIME LAB |
| Well 57/ | 4809 ± 342[†] | - |
| QA-1 | 4303 ± 236 | - |
| Well 36/ | 21477 ± 1439 | - |
| QA-2 | 23451 ± 493 | - |
| Well 38/ | 3655 ± 225 | - |
| QA-3 | 4440 ± 195 | - |
| CPP-4/ | 1792 ± 42 | - |
| QA-4 | 1954 ± 138 | - |
| Well 67/ | 2841 ± 124 | - |
| QA-5 | 2733 ± 137 | - |
| Well 11 | 1670 ± 110 | - |
| | 1689 ± 104 | - |
| Well 14 | 2030 ± 100 | - |
| | 2017 ± 105 | - |
| CFA-1 | 2164 ± 105 | 2090 ± 170 |
| Well 39 | 1207 ± 89 | 1089 ± 74 |
| Well 41 | 536 ± 29 | 567 ± 35 |
| Well 53 | - | 212 ± 20 |
| | - | 201 ± 26 |
| Well 54 | - | 65 ± 9 |
| | - | 74 ± 5 |
| Well 68 | - | 40 ± 6 |
| | - | 35 ± 5 |

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[†] Uncertainties represent the 67 percent confidence interval.

# APPENDIX 2

## RESULTS FROM ON-SITE WELLS[†] INFLUENCED BY WASTE DISPOSAL PRACTICES AT THE IDAHO NATIONAL ENGINEERING LABORATORY

| Sample | Chloride (ppm) | *In-Situ* $^{36}$Cl/Cl ($10^{-15}$) | Atoms $^{36}$Cl liter$^{-1}$ ($10^{10}$) |
|---|---|---|---|
| NRF-1 | 34 | 1125 ± 87 | 0.065 ± 0.005[§] |
| NRF-2 | 41 | 937 ± 131 | 0.065 ± 0.009 |
| NRF-3 | 35 | 998 ± 75 | 0.059 ± 0.004 |
| Well 97 | 33 | 1046 ± 47 | 0.059 ± 0.003 |
| Well 99 | 20 | 881 ± 162 | 0.027 ± 0.006 |
| Site 4 | 26 | 2270 ± 170 | 0.100 ± 0.007 |
| Fire Station 2 | 16 | 997 ± 67 | 0.030 ± 0.002 |
| Site 19 | 11 | 539 ± 43 | 0.010 ± 0.001 |
| TRA-Disposal | 12 | 100000 ± 5000 | 2.0 ± 0.1 |
| MTR Test | 13 | 35500 ± 5000 | 0.78 ± 0.11 |
| Well 121 | 14 | 2540 ± 220 | 0.060 ± 0.005 |
| Well 65 | 21 | 1560000 ± 94000 | 56 ± 3 |
| Well 58 | 12 | 125000 ± 5000 | 2.5 ± 0.1 |
| ICPP-4 | 17 | 1792 ± 42 | 0.052 ± 0.001 |
| ICPP-2 | 17 | 5602 ± 198 | 0.16 ± 0.01 |
| ICPP-1 | 16 | 45419 ± 1317 | 1.2 ± 0.04 |
| NPR Test | 20 | 1231 ± 165 | 0.042 ± 0.006 |
| Well 43 | 37 | 1580000 ± 80000 | 99 ± 5 |
| Well 50 | 80 | 2790000 ± 73500 | 380 ± 10 |
| Well 40 | 28 | 547000 ± 21000 | 26 ± 1 |
| Well 46 | 24 | 319000 ± 17000 | 13 ± 0.7 |
| Well 44 | 18 | 49200 ± 3300 | 1.5 ± 0.1 |
| Well 52 | 30 | 1890000 ± 39000 | 96 ± 2 |
| Well 41 | 24 | 540000 ± 24500 | 22 ± 1 |
| Well 45 | 21 | 87000 ± 4600 | 3.1 ± 0.2 |
| Well 42 | 21 | 155000 ± 14000 | 5.5 ± 0.5 |
| Well 47 | 28 | 489000 ± 32000 | 23 ± 2 |
| Well 48 | 23 | 385000 ± 25000 | 15 ± 1 |
| Well 82 | 19 | 180000 ± 9000 | 5.8 ± 0.3 |
| Well 39 | 11 | 267000 ± 21000 | 5.0 ± 0.4 |
| Well 59 | 45 | 262000 ± 13000 | 20 ± 1 |
| Well 35 | 24 | 1120000 ± 50000 | 46 ± 2 |
| Well 51 | 60 | 2180000 ± 39000 | 222 ± 4 |
| Well 67 | 82 | 835000 ± 36000 | 116 ± 5 |
| Well 34 | 18 | 840000 ± 30000 | 26 ± 1 |
| Well 57 | 76 | 1550000 ± 110000 | 200 ± 14 |
| Well 36 | 39 | 1360000 ± 90000 | 90 ± 6 |

## APPENDIX 2 (Cont'd)

| Sample | Chloride (ppm) | *In-Situ* $^{36}$Cl/Cl ($10^{-15}$) | Atoms $^{36}$Cl liter$^{-1}$ ($10^{10}$) |
|---|---|---|---|
| Well 37 | 59 | 1200000 ± 50000 | 120 ± 5 |
| Well 114 | 80 | 800000 ± 29000 | 109 ± 4 |
| Well 111 | 103 | 366000 ± 11000 | 64 ± 2 |
| Well 115 | 35 | 455000 ± 17000 | 27 ± 1 |
| Well 116 | 71 | 475000 ± 17000 | 57 ± 2 |
| Well 38 | 118 | 750000 ± 45000 | 150 ± 9 |
| Well 77 | 115 | 650000 ± 31000 | 127 ± 6 |
| Well 112 | 96 | 976000 ± 39000 | 159 ± 6 |
| Well 113 | 145 | 549000 ± 12000 | 135 ± 3 |
| Well 85 | 44 | 1490000 ± 10000 | 111 ± 1 |
| Well 20 | 24 | 900000 ± 73000 | 37 ± 3 |
| CFA - 1 | 98 | 482000 ± 25000 | 80 ± 4 |
| Site 9 | 12 | 311 ± 18 | 0.006 ± 0.0004 |
| Badge Facility | 17 | 1744 ± 87 | 0.050 ± 0.003 |
| EBR - 1 | 7 | 1547 ± 235 | 0.018 ± 0.003 |
| Well 83 | 12 | 468 ± 21 | 0.010 ± 0.0005 |
| Well 107 | 20 | 338 ± 17 | 0.011 ± 0.0006 |
| Well 87 | 14 | 177000 ± 5100 | 4.2 ± 0.1 |
| RWMC Production | 15 | 227000 ± 19000 | 5.8 ± 0.5 |
| Well 117 | 14 | 23300 ± 1000 | 0.55 ± 0.02 |
| Well 90 | 14 | 216000 ± 6300 | 5.1 ± 0.2 |
| Well 88 (s) | 90 | 428 ± 47 | 0.065 ± 0.007[1] |
| Well 88 (h) | 84 | 410 ± 21 | 0.058 ± 0.003[1] |
| Well 120 | 26 | 11800 ± 590 | 0.52 ± 0.03 |
| Well 106 | 15 | 274000 ± 7100 | 7.0 ± 0.2 |
| Well 104 | 12 | 279000 ± 5000 | 5.7 ± 0.1 |
| Well 9 | 23 | 2700 ± 314 | 0.11 ± 0.01 |
| Well 109 | 16 | 1480 ± 74 | 0.040 ± 0.002 |
| Well 105 | 15 | 29900 ± 1200 | 0.76 ± 0.03 |
| Well 108 | 15 | 21700 ± 600 | 0.55 ± 0.02 |
| Well 103 | 16 | 1434 ± 70 | 0.039 ± 0.002 |
| Well 110 | 19 | 338 ± 17 | 0.011 ± 0.0006 |
| Well 11 | 14 | 1680 ± 151 | 0.040 ± 0.004[2] |
| Well 14 | 21 | 2024 ± 145 | 0.073 ± 0.005[2] |

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[†] Wells completed in the Snake River Plain aquifer. Wells 11 and 14 lie approximately 6 and 12 km from the southern site boundary.

[§] Uncertainties represent 67 percent confidence intervals.

[1] Sample 88(h) was collected with a piston pump; 88(s) with a submersible pump. Sampling was done on the same day.

[2] Average of duplicates.

Conversion factors: $10^{10}$ atoms $l^{-1}$ = 0.02 pCi $l^{-1}$

# APPENDIX 3

## RESULTS FROM ON-SITE WELLS[‡] UNINFLUENCED BY RELEASES FROM THE TEST REACTOR AREA (TRA) AND THE IDAHO CHEMICAL PROCESSING PLANT (ICPP)

| Sample | Chloride (ppm) | *In Situ* $^{36}$Cl/Cl ($10^{-15}$) | Atoms $^{36}$Cl liter$^{-1}$ ($10^{10}$) |
|---|---|---|---|
| Site 17 | 11 | 622 ± 93 | 0.012 ± 0.002[§] |
| Arbor Test | 13 | 367 ± 22 | 0.008 ± 0.0005 |
| Well 2 | 16 | 377 ± 19 | 0.010 ± 0.0005 |
| Well 101 | 8 | 255 ± 14 | 0.004 ± 0.0002 |
| Well 100 | 16 | 398 ± 20 | 0.011 ± 0.0005 |
| Site 9 | 12 | 311 ± 18 | 0.006 ± 0.0004 |
| Well 86 | 23 | 447 ± 44 | 0.017 ± 0.0020 |
| Well 7 | 10 | 113 ± 13 | 0.002 ± 0.0002 |
| Well 23 | 11 | 350 ± 29 | 0.007 ± 0.0006 |
| Well 29 | 29 | 417 ± 80 | 0.021 ± 0.0040 |
| Well 12 | 33 | 916 ± 40 | 0.051 ± 0.0022 |
| P&W 2 | 8 | 375 ± 46 | 0.005 ± 0.0006 |
| Well 19 | 11 | 539 ± 43 | 0.010 ± 0.0008 |
| Site 14 | 9 | 287 ± 69 | 0.004 ± 0.0010 |
| Well 32 | 59 | 282 ± 17 | 0.028 ± 0.0017 |
| Mean | | 404 ± 185[1] | 0.013 ± 0.013[†] |

[‡] Wells completed in the Snake River Plain aquifer.

[§] Uncertainties represent 67 percent confidence interval.

[†] Standard Deviation (1$\sigma$; n = 14).

Conversion factors: $10^{10}$ atoms $l^{-1}$ = 0.02 pCi $l^{-1}$

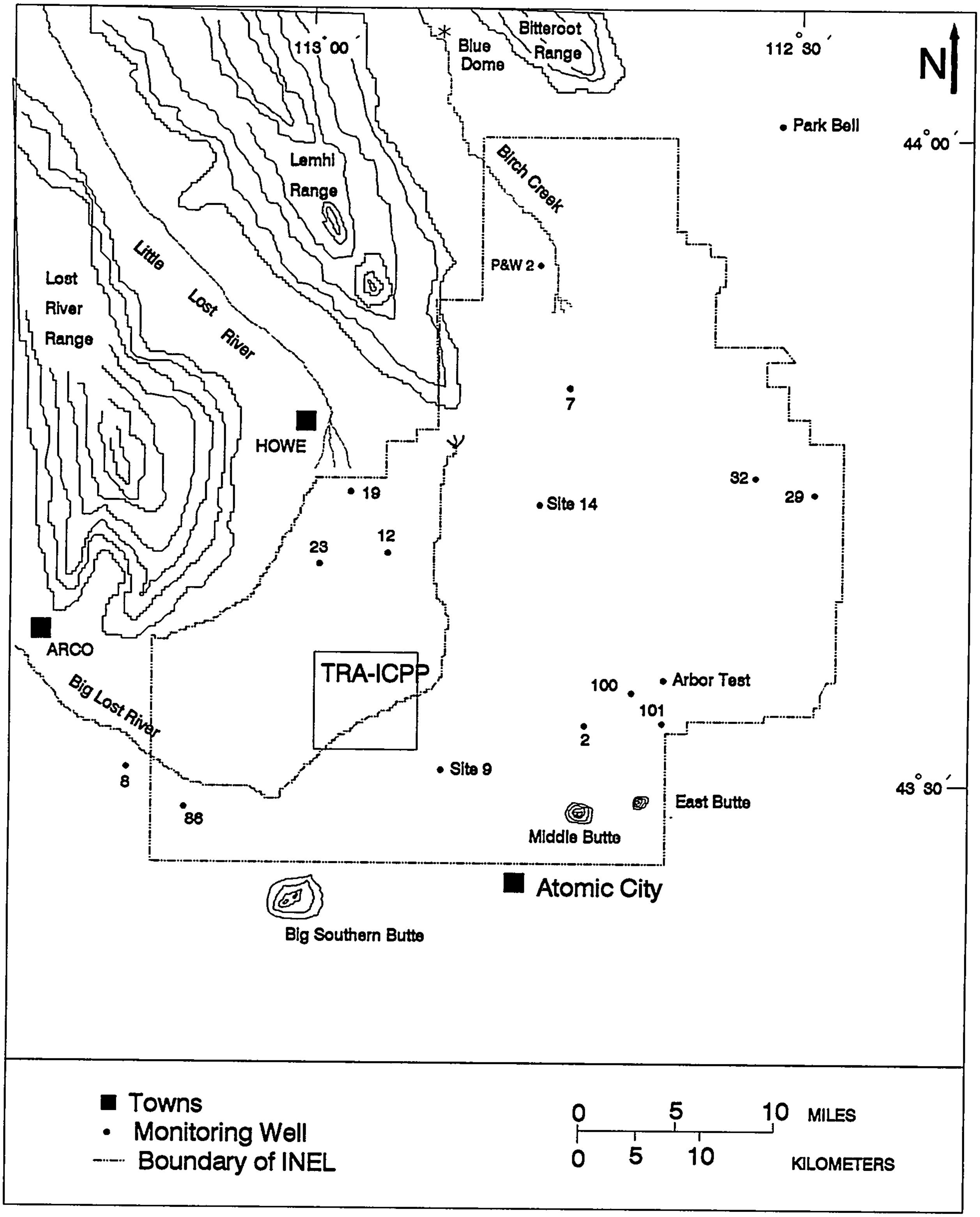

Figure 10. Wells uninfluenced by $^{36}$Cl plume originating at the TRA and the ICPP.

# APPENDIX 4

## RESULTS FOR OFF-SITE WELLS AND SURFACE WATER

| Sample | Chloride (ppm) | *In-Situ* $^{36}$Cl/Cl ($10^{-15}$) | Atoms $^{36}$Cl liter$^{-1}$ ($10^{10}$) |
|---|---|---|---|
| Park Bell | 6 | 213 ± 28 | 0.002 ± 0.0003 |
| Well 8 | 9 | 817 ± 41 | 0.012 ± 0.0006 |
| Arco City | 6 | 2199 ± 82[§] | 0.022 ± 0.0008[1] |
| Arco City | 5 | 2353 ± 163 | 0.020 ± 0.0014[1] |
| Ruby Farms Well | 46 | 584 ± 40 | 0.046 ± 0.0032[2] |
| Ruby Farms Well | 25 | 587 ± 55 | 0.025 ± 0.0023[2] |
| Big Lost River | 4 | 2461 ± 99 | 0.017 ± 0.0007[3] |
| Big Lost River | 3 | 2776 ± 168 | 0.014 ± 0.0008[3] |
| East Fork Big Lost River | 2 | 5257 ± 247 | 0.018 ± 0.0008 |
| Birch Creek | 5 | 623 ± 33 | 0.005 ± 0.0003[4] |
| Birch Creek | 4 | 607 ± 36 | 0.004 ± 0.0002[5] |
| Little Lost River Springs | 13 | 1519 ± 157 | 0.033 ± 0.0035[6] |
| Upper Little Lost River | 3 | 3256 ± 263 | 0.017 ± 0.0013[6] |
| Camus Creek | 8 | 1251 ± 97 | 0.017 ± 0.0014[6] |
| Star Hope Creek | 0.64 | 9779 ± 819 | 0.011 ± 0.0009[6] |
| Mean | | | 0.018 ± 0.011[†] |

[§] Uncertainties represent 67 percent confidence interval.

[1] Arco City Municipal Well collected in 1990/1991, respectively.

[2] Ruby Farms Well near Howe, ID collected 1990/1991, respectively.

[3] Big Lost River at Mackay, ID collected 1990/1991, respectively.

[4] Birch Creek at Blue Dome Inn, Blue Dome, ID (1990)

[5] Birch Creek at Kaufman Compound (1991).

[6] Collected in 1991. These sampling sites are not shown in any figure; they all lie at distances > 100 km from the INEL.

[†] Standard Deviation ($1\sigma$; n = 15).

Conversion factors: $10^{10}$ atoms $l^{-1}$ = 0.02 pCi $l^{-1}$

# APPENDIX 5

## RESULTS FOR THE EASTERN SNAKE RIVER PLAIN AQUIFER DOWNGRADIENT FROM THE IDAHO NATIONAL ENGINEERING LABORATORY

| Sample | Chloride (ppm) | *In-Situ* $^{36}Cl/Cl$ ($10^{-15}$) | Atoms $^{36}Cl$ liter$^{-1}$ ($10^{10}$) |
|---|---|---|---|
| Well 14 | 21.0 | 2150 ± 50[§] | 0.077 ± 0.002[†] |
| Houghland Well | 13.6 | 609 ± 18 | 0.014 ± 0.0004 |
| BLM Grazing Well | 11.9 | 267 ± 8 | 0.005 ± 0.0002 |
| Crossroads Well | 9.1 | 1006 ± 20 | 0.016 ± 0.0003 |
| Finger Buttes Well | 9.8 | 227 ± 7 | 0.004 ± 0.0001 |
| CCC-3 Well | 12.9 | 283 ± 8 | 0.006 ± 0.0002 |
| MV-1 | 48.3 | 257 ± 9 | 0.021 ± 0.0007 |
| MV-4 | 57.4 | 223 ± 8 | 0.022 ± 0.0008 |
| MV-9 | 100.4 | 320 ± 9 | 0.055 ± 0.0015 |
| MV-11 | 69.4 | 347 ± 12 | 0.041 ± 0.0014 |
| MV-12 | 62.8 | 285 ± 13 | 0.003 ± 0.0001 |
| MV-14 | 47.5 | 289 ± 15 | 0.023 ± 0.0012 |
| MV-15 | 47.3 | 348 ± 16 | 0.028 ± 0.0013 |
| MV-18 | 54.1 | 345 ± 10 | 0.032 ± 0.0009 |
| MV-21 | 10.5 | 397 ± 25 | 0.007 ± 0.0004 |
| MV-23 | 47.0 | 324 ± 11 | 0.026 ± 0.0009 |
| MV-30 | 52.6 | 385 ± 10 | 0.034 ± 0.0009 |
| MV-33 | 7.1 | 391 ± 11 | 0.005 ± 0.0001 |
| MV-37 | 22.0 | 867 ± 18 | 0.032 ± 0.0007 |
| MV-39 | 29.3 | 691 ± 34 | 0.034 ± 0.0016 |
| MV-43 | 46.4 | 409 ± 11 | 0.032 ± 0.0009 |
| MV-51 | 55.4 | 349 ± 11 | 0.033 ± 0.0010 |
| MV-59 | 13.1 | 139 ± 9 | 0.003 ± 0.0002 |

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[†] Remeasured in 1994; see Appendix 2 for 1992 value.

[§] Uncertainties represent 67 percent confidence intervals

Conversion factors: $10^{10}$ atoms $l^{-1}$ = 0.02 pCi $l^{-1}$

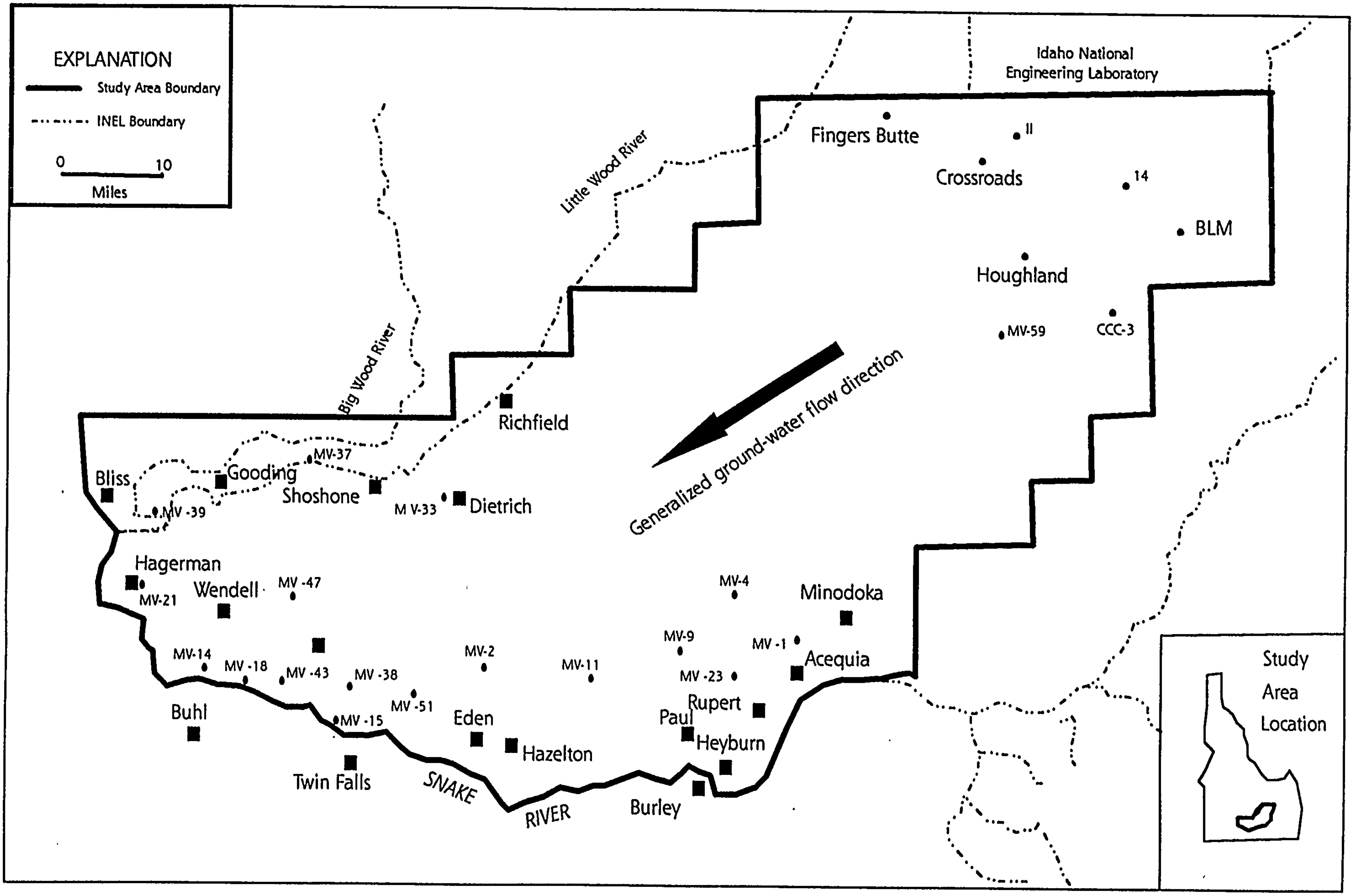

Figure 11. Wells in the eastern Snake River Plain downgradient from the Idaho National Engineering Laboratory. All streams, rivers, and tributaries to the Snake River are not shown.

# APPENDIX 6

## RESULTS FOR WELLS AT THE TEST AREA NORTH (TAN)

| Sample | Chloride (ppm) | *In-Situ* $^{36}$Cl/Cl ($10^{-15}$) | Atoms $^{36}$Cl liter$^{-1}$ ($10^{10}$) |
|---|---|---|---|
| TAN-1 | 15.8 | 842 ± 35§ | 0.023 ± 0.0010 |
| TAN-2 | 12.4 | 1631 ± 55 | 0.034 ± 0.0011 |
| TAN-3 | 11.4 | 685 ± 89 | 0.013 ± 0.0017 |
| TAN-4 | 28.6 | 5840 ± 240 | 0.284 ± 0.012 |
| TAN-5 | 30.5 | 4433 ± 194 | 0.230 ± 0.010 |
| TAN-6 | 7.4 | 242 ± 12 | 0.0030 ± 0.0001 |
| TAN-7 | 11.4 | 467 ± 21 | 0.0090 ± 0.0004 |
| TAN-8 | 40.6 | 324 ± 17 | 0.022 ± 0.0011 |
| TAN-9 | 116.8 | 1520 ± 290 | 0.302 ± 0.058 |
| TAN-10A | 106.7 | 437 ± 18 | 0.079 ± 0.003 |
| TAN-11 | 49.7 | 3250 ± 140 | 0.274 ± 0.012 |
| TAN-12 | 42.2 | 1530 ± 60 | 0.110 ± 0.004 |
| TAN-13A | 3.4 | 288 ± 24 | 0.0017 ± 0.0001 |
| TAN-14 | 3.6 | 257 ± 25 | 0.0016 ± 0.0002 |
| TAN-15 | 20.0 | 1983 ± 47 | 0.067 ± 0.002 |
| TAN-16 | 17.9 | 2088 ± 96 | 0.064 ± 0.003 |
| TAN-17 | 4.4 | 308 ± 25 | 0.002 ± 0.0002 |
| TAN-24 | 71.3 | 3690 ± 150 | 0.447 ± 0.018 |
| TAN Disposal-1 | 43.2 | 4910 ± 25 | 0.360 ± 0.002 |
| TAN Disposal-2 | 109.6 | 1040 ± 30 | 0.194 ± 0.006 |
| TAN Disposal-3 | 13.5 | 540 ± 30 | 0.012 ± 0.001 |
| FET-2 | 16.7 | 376 ± 24 | 0.011 ± 0.001 |
| GIN-2 | 45.4 | 8730 ± 200 | 0.673 ± 0.015 |
| GIN-4 | 47.7 | 10000 ± 400 | 0.810 ± 0.032 |
| ANP-6 | 11.5 | 290 ± 19 | 0.0057 ± 0.0004 |

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§ Uncertainties represent 67 percent confidence intervals.
Conversion factors: $10^{10}$ atoms $l^{-1}$ = 0.02 pCi $l^{-1}$

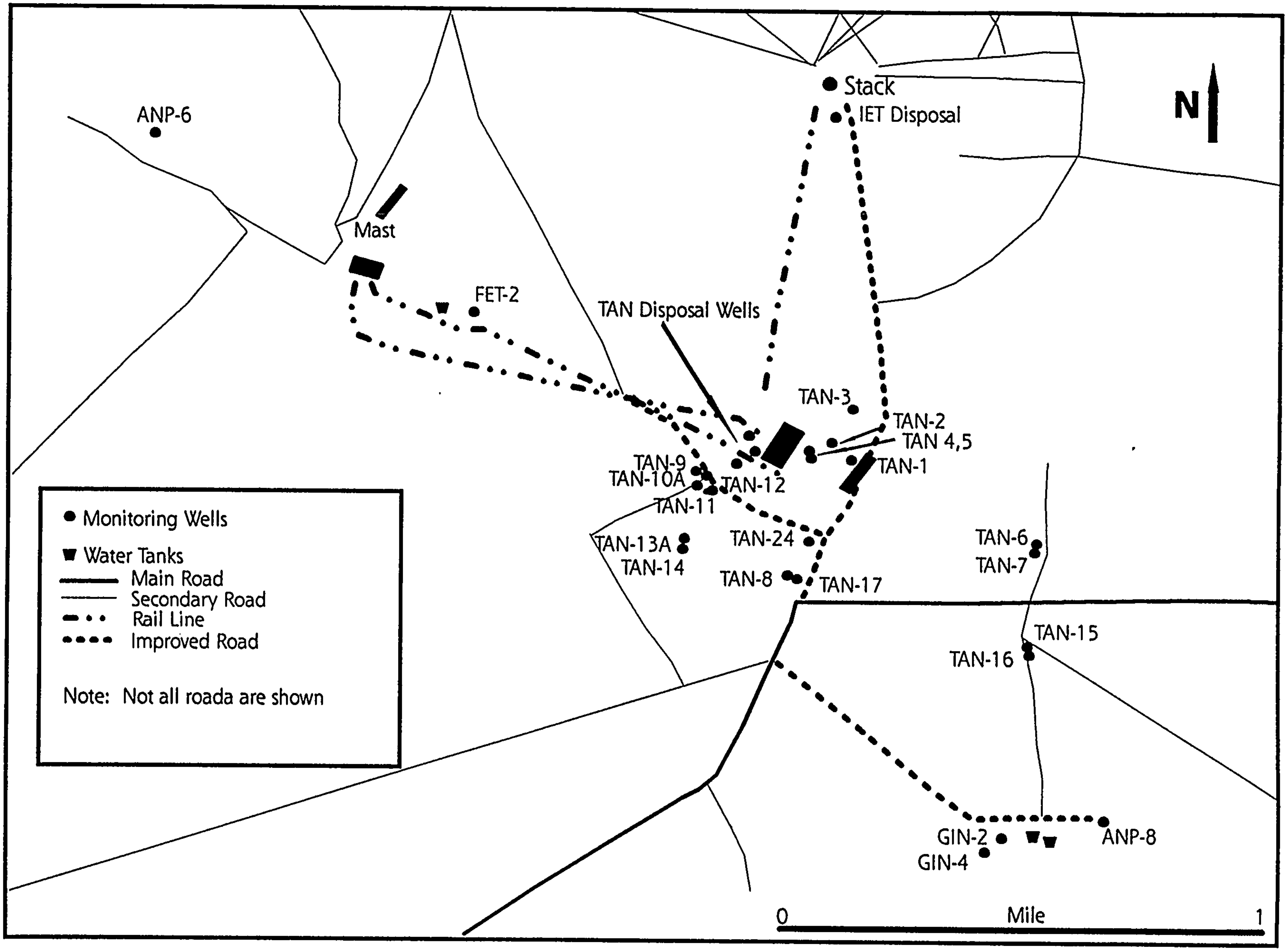

Figure 12. Wells sampled in Test Area North (TAN). Gas injection wells (GIN-2,4) show highest levels of $^{36}$Cl.

# APPENDIX 7

## RESULTS FROM WELLS DRAWING PERCHED WATER IN THE TEST REACTOR AREA (TRA)

| Sample | Chloride (ppm) | *In Situ* $^{36}$Cl/Cl ($10^{-15}$) | Atoms $^{36}$Cl liter$^{-1}$ ($10^{10}$) |
|---|---|---|---|
| Well 68 | 44 | 2100 ± 500[§] | 0.16 ± 0.04[1] |
| Well 56 | 10 | 55300 ± 2900 | 0.94 ± 0.05 |
| Well 55 | 29 | 2880000 ± 282000 | 142 ± 14[2] |
| Well 53 | 18 | 27700 ± 4500 | 0.85 ± 0.14[1] |
| Well 54 | 22 | 7800 ± 1100 | 0.29 ± 0.04[1] |
| Well 63 | 20 | 20300 ± 2400 | 0.69 ± 0.08 |
| Well 60 | 21 | 3300 ± 500 | 0.12 ± 0.02 |
| Well 70 | 20 | 115000 ± 8800 | 3.9 ± 0.3 |
| Well 69 | 20 | 6800 ± 900 | 0.23 ± 0.03 |
| Well 61 | 18 | 142000 ± 9700 | 4.3 ± 0.3 |
| Well 62 | 21 | 27800 ± 1700 | 1.0 ± 0.06 |

[§] Uncertainties represent 67 percent confidence interval.

[1] Average of duplicate analyses (see Appendix 1).

[2] In calculating an average $^{36}$Cl atom concentration in the perched zone(see text under Results and Discission), the value for Well 55 was omitted. Although the measured atom concentration is believed to be real, it is doubtful that the areal extent of such highly labeled water is significant, based on data from the surrounding wells as shown.

Conversion factors: $10^{10}$ atoms l$^{-1}$ = 0.02 pCi l$^{-1}$

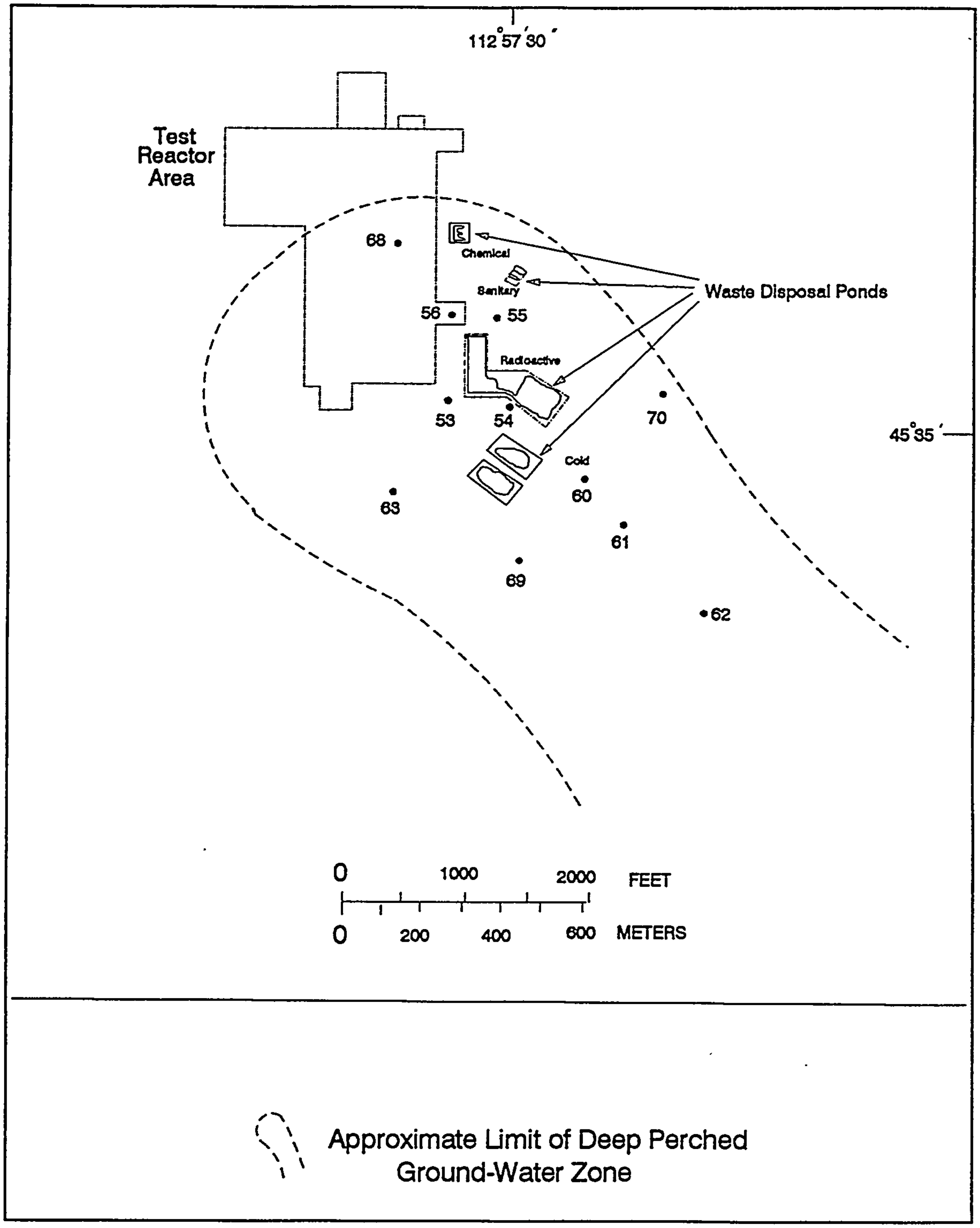

Figure 13. Wells in Test Reactor Area that sample deep, perched-water zone. (adapted from Pittman et al., 1988).