

Preliminary Delineation of Natural Geochemical Reactions, Snake River Plain Aquifer System, Idaho National Engineering Laboratory and Vicinity, Idaho

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CONVERSION FACTORS, VERTICAL DATUM, AND ABBREVIATED UNITS

	Multiply	By	To Obtain
	inch (in.)	25.4	millimeter
	foot (ft)	0.3048	meter
	mile (mi)	1.609	kilometer
	acre	0.4047	hectare
	square mile (mi ²)	2.590	square kilometer
	gallon (gal)	3.785	liter
	acre-foot (acre-ft)	1,233	cubic meter
	foot per mile (ft/mi)	0.1894	meter per kilometer
	cubic foot per second per mile ((ft ³ /s)/mi)	0.01760	cubic meter per second per kilometer
	pound (lb)	0.4536	kilogram
	curie (Ci)	3.7x10 ¹⁰	becquerel
	picocurie per milliliter (pCi/mL)	0.037	becquerel per milliliter
	picocurie per liter (pCi/L)	0.037	becquerel per liter

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

Sea level: In this report, "sea level" refers to the National Geodetic Vertical Datum of 1929—a geodetic datum derived from a general adjustment of the first-order level nets of both the United States and Canada, formerly called Sea Level Datum of 1929.

Abbreviated units used in report: $\mu\text{g/L}$ (microgram per liter), mg/L (milligram per liter), and ppm (parts per million).

PRELIMINARY DELINEATION OF NATURAL GEOCHEMICAL REACTIONS, SNAKE RIVER PLAIN AQUIFER SYSTEM, IDAHO NATIONAL ENGINEERING LABORATORY AND VICINITY, IDAHO

By LeRoy L. Knobel, Roy C. Bartholomay, and Brennon R. Orr

Abstract

The U.S. Geological Survey, in cooperation with the U.S. Department of Energy, is conducting a study to determine the natural geochemistry of the Snake River Plain aquifer system at the Idaho National Engineering Laboratory (INEL), Idaho. As part of this study, a group of geochemical reactions that partially control the natural chemistry of ground water at the INEL were identified. Mineralogy of the aquifer matrix was determined using X-ray diffraction and thin-section analysis and theoretical stabilities of the minerals were used to identify potential solid-phase reactants and products of the reactions. The reactants and products that have an important contribution to the natural geochemistry include labradorite, olivine, pyroxene, smectite, calcite, ferric oxyhydroxide, and several silica phases.

To further identify the reactions, analyses of 22 representative water samples from sites tapping the Snake River Plain aquifer system were used to determine the thermodynamic condition of the ground water relative to the minerals in the framework of the aquifer system. Principal reactions modifying the natural geochemical system include congruent dissolution of olivine, diopside, amorphous silica, and anhydrite; incongruent dissolution of labradorite with calcium montmorillonite as a residual product; precipitation of calcite and

ferric oxyhydroxide; and oxidation of ferrous iron to ferric iron. Cation exchange reactions retard the downward movement of heavy, multivalent waste constituents where infiltration ponds are used for waste disposal.

INTRODUCTION

The Idaho National Engineering Laboratory (INEL), encompassing about 890 mi² of the eastern Snake River Plain in southeastern Idaho (fig. 1), is operated by the U.S. Department of Energy (DOE). INEL facilities are used in the development of peacetime atomic-energy applications, nuclear safety research, defense programs, and advanced energy concepts. Liquid radionuclide and chemical wastes generated at these facilities have been discharged to onsite infiltration ponds and disposal wells since 1952. Liquid-waste disposal has resulted in detectable concentrations of several waste constituents in water in the Snake River Plain aquifer underlying the INEL.

The DOE requires information about the mobility of dilute radionuclide- and chemical-waste constituents in the Snake River Plain aquifer. Waste-constituent mobility is, in part, determined by (1) the rate and direction of ground-water flow; (2) the locations, quantities, and methods of waste disposal; (3) waste-constituent chemistry; and (4) the geochemical

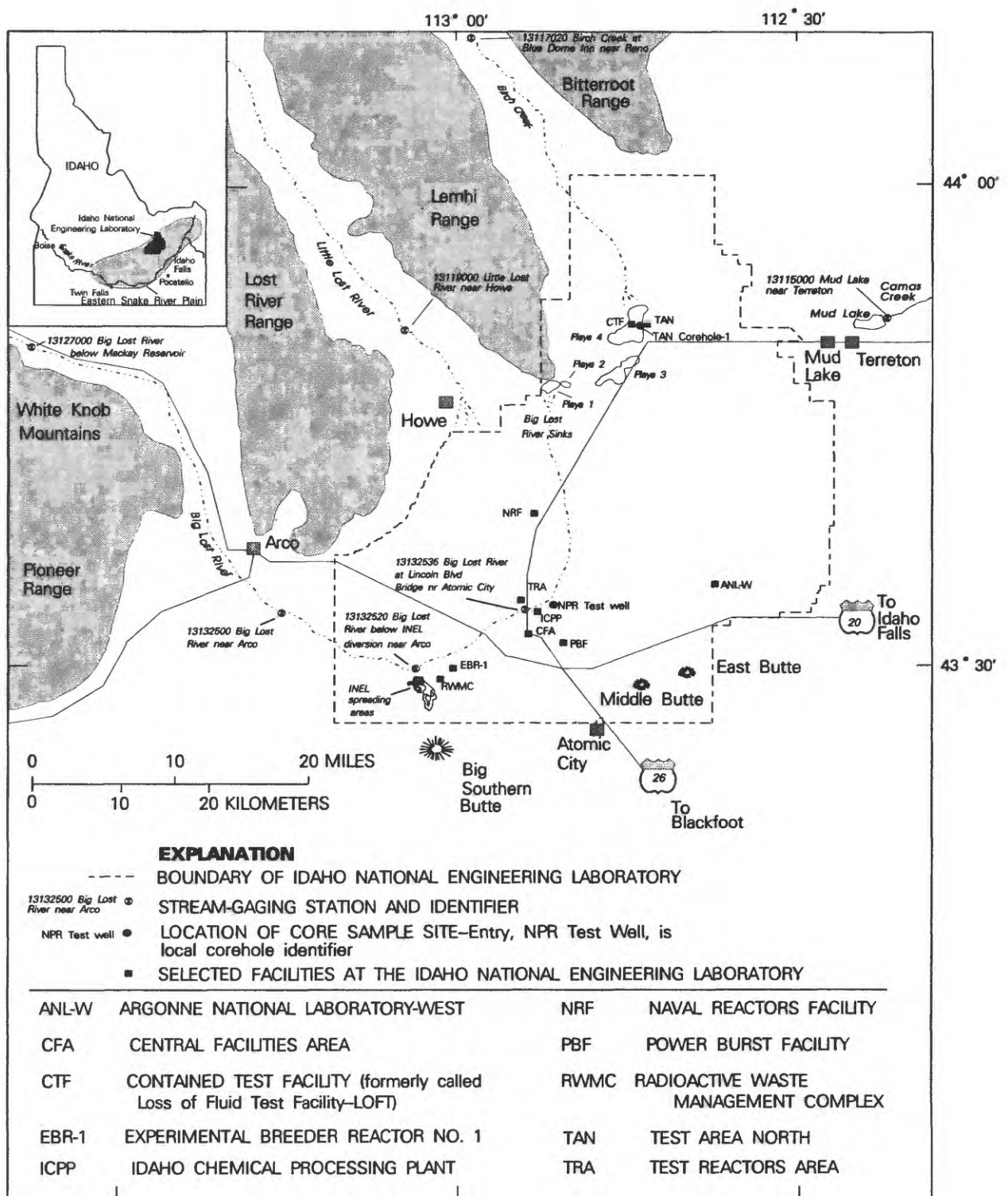


Figure 1. Location of the Idaho National Engineering Laboratory, stream-gaging stations, and selected facilities.

processes taking place in the aquifer. This study was conducted by the U.S. Geological Survey (USGS) in cooperation with the DOE's Idaho Operations Office.

Purpose and Scope

In 1949, the U.S. Atomic Energy Commission, later to become the DOE, requested that the USGS describe the water resources of the area now known as the INEL. The purpose of the resulting study was to characterize these resources prior to the development of nuclear reactor testing facilities. The USGS since has maintained a monitoring network at the INEL to determine hydrologic trends and to delineate the movement of facility-related radiochemical and chemical wastes in the Snake River Plain aquifer.

This report presents a preliminary delineation of natural geochemical reactions occurring in the Snake River Plain aquifer system as part of the continuing hydrogeologic investigation at the INEL. The report describes the mineralogy, mineral stability, ion distribution, hydrochemical facies, and thermodynamic condition of the aquifer system. A companion report dealing with the quantitative aspects of the natural geochemical system at the INEL will be published after additional data are obtained.

Waste Disposal at the INEL

Liquid-waste disposal sites at INEL facilities have been the principal sources of radiochemical- and chemical-waste constituents in water from the Snake River Plain aquifer at the INEL. Liquid-waste disposal sites at the INEL have included infiltration ponds and ditches, drain fields, and disposal wells. Waste materials buried at the Radioactive Waste Management Complex (RWMC) also are a source of some waste constituents in

ground water. Radiochemical waste-disposal data presented in this report were obtained from a series of radioactive waste-management information reports (Litteer and Reagan, 1990; Litteer and others, 1991b; Litteer and others, 1993). Chemical waste-disposal data were obtained from a series of industrial waste-management information reports (Litteer and Peterson, 1990; Litteer and others, 1991a; Litteer and others, 1992). The radiochemical- and chemical-waste disposal data are collected by contractors at each facility.

Acknowledgments

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HYDROLOGIC CONDITIONS

The Snake River Plain aquifer is one of the most productive aquifers in the United States (U.S. Geological Survey, 1985, p. 193). The aquifer consists of a thick sequence of basalts and sedimentary interbeds filling a large, arcuate, structural basin that underlies the eastern Snake River Plain in southeastern Idaho (fig. 1).

Recharge to the Snake River Plain aquifer principally is from infiltration of applied irrigation water, infiltration of streamflow, and alluvial ground-water inflow from adjoining mountain drainage basins. Some recharge may be from direct infiltration of precipitation, although the small annual precipitation on the plain (8 in. at the INEL), evapotranspiration, and the great depth to water (in places exceeding 900 ft) probably minimize this source of recharge.

Surface Water

The Big Lost River drains more than 1,400 mi² of mountainous area that includes parts of the Lost River Range and Pioneer Range west of the INEL (fig. 1). Flow in the Big Lost River infiltrates to the Snake River Plain aquifer along its channel and at sinks and playas at the river's terminus. Since 1958, excess runoff has been diverted to spreading areas in the southwestern part of the INEL where much of the water rapidly infiltrates to the aquifer. Other surface drainages that provide recharge to the Snake River Plain aquifer at the INEL include Birch Creek and the Little Lost River (fig. 1).

The average streamflow in the Big Lost River below Mackay Reservoir for the 75-year period of record (water years, 1905, 1913-14, and 1920-91) was 223,900 acre-ft/year (Harenberg and others, 1992, p. 179). Streamflow in the Big Lost River below Mackay Reservoir increased from 135,200 acre-ft during the 1989 water year (60 percent of average flow) to 165,100 acre-ft during the 1991 water year (74 percent of average flow) (fig. 2). No streamflow was recorded in the Big Lost River near Arco (fig. 1), in the Big Lost River below the INEL diversion near Arco (fig. 2), and the INEL diversion at its head near Arco (fig. 2) during the 1989-91 water years.

Recharge to the Snake River Plain aquifer can be substantial downstream from Arco because of infiltration through the Big Lost River channel, diversion areas, sinks, and playas. Measured infiltration losses at various discharges ranged from 1 to 28 (ft³/s)/mi (Bennett, 1990, p. 1).

Ground Water

Water in the Snake River Plain aquifer moves principally through fractures and interflow zones in the basalt. A significant proportion of ground water moves through the

upper 800 ft of saturated rocks (Mann, 1986, p. 21). Hydraulic conductivity of basalt in the upper 800 ft of the aquifer generally is 1 to 100 ft/day. Hydraulic conductivity of underlying rocks is several orders of magnitude smaller.

Depth to water in wells completed in the Snake River Plain aquifer ranges from about 200 ft in the northern part of the INEL to more than 900 ft in the southeastern part. In July 1991, the altitude of the water table (fig. 3) was about 4,585 ft above sea level near TAN (fig. 1) and about 4,425 ft above sea level near the RWMC (fig. 1). Water flowed southward and southwestward beneath the INEL at an average hydraulic gradient of about 4 ft/mi (fig. 3). Locally, however, the hydraulic gradient ranged from about 1 to 15 ft/mi. From July 1988 to July 1991, water levels generally declined throughout the INEL because of drought conditions that began in 1987. Water-level declines ranged from 10 ft in a well north of the Naval Reactors Facility (NRF) to about 2 ft in wells located in the southern part of the site (figs. 1, 4). The greater water-level decline in the well north of the NRF is attributed to lack of recharge from the Big Lost River during the drought period.

Ground water moves southwestward from the INEL and eventually is discharged to springs along the Snake River downstream from Twin Falls, 100 mi southwest of the INEL. Approximately 4.3 million acre-ft of ground water were discharged to these springs in 1988 (Mann, 1989, p. 2).

GEOCHEMICAL CONDITIONS

Mineralogy and Mineral Stability

Mineralogy of extrusive igneous rocks is dependent on the chemical composition of the parent magma and on the history of temperature and pressure changes in the magma during its ascent to the Earth's surface.

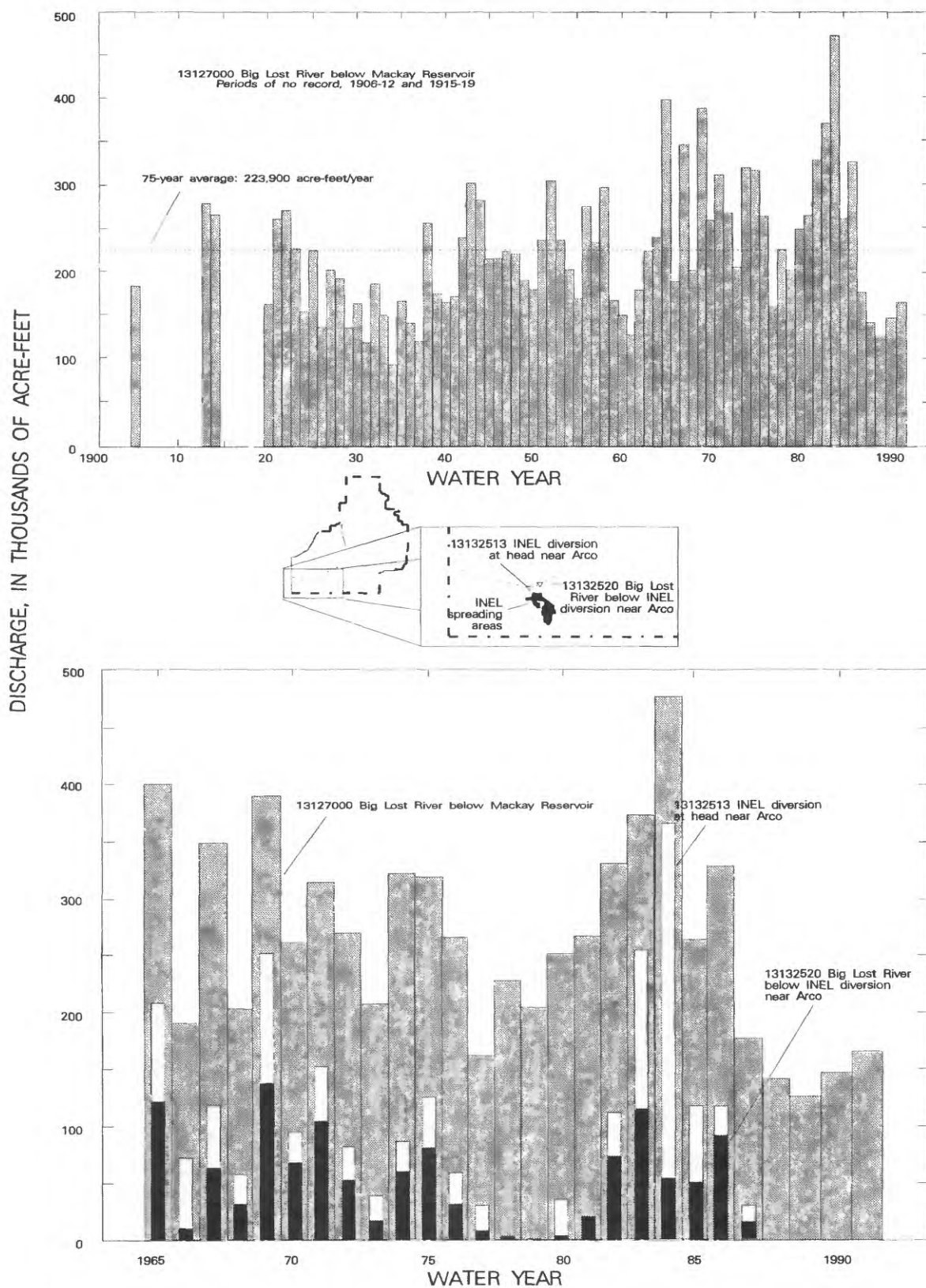


Figure 2. Discharge of the Big Lost River below Mackay Reservoir (water years 1905, 1913–14, and 1920–91), Big Lost River below the INEL diversion near Arco, and the INEL diversion at head near Arco (water years 1965–91).

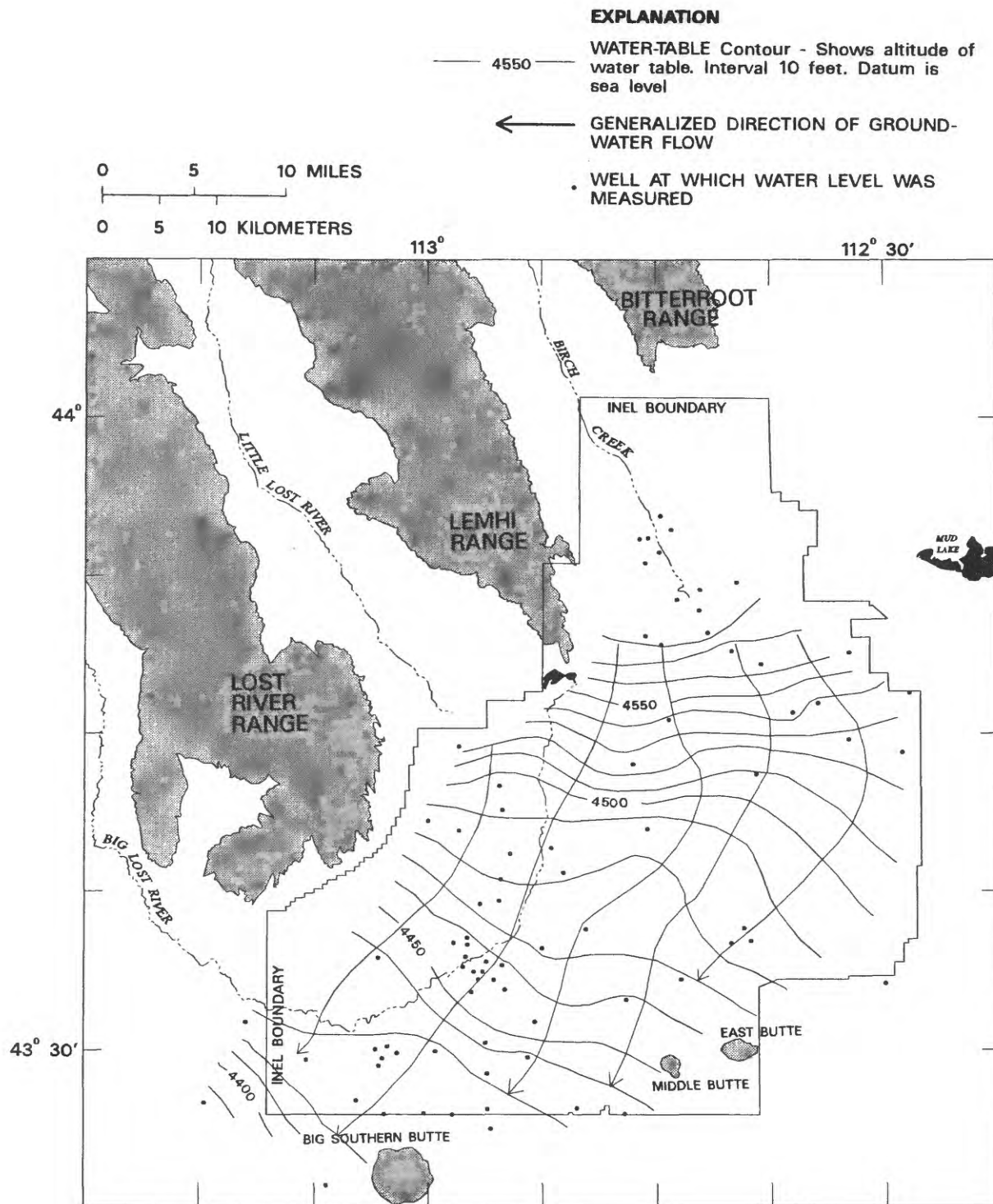


Figure 3. Altitude of the water table, Snake River Plain aquifer, and general direction of ground-water movement, July 1991.

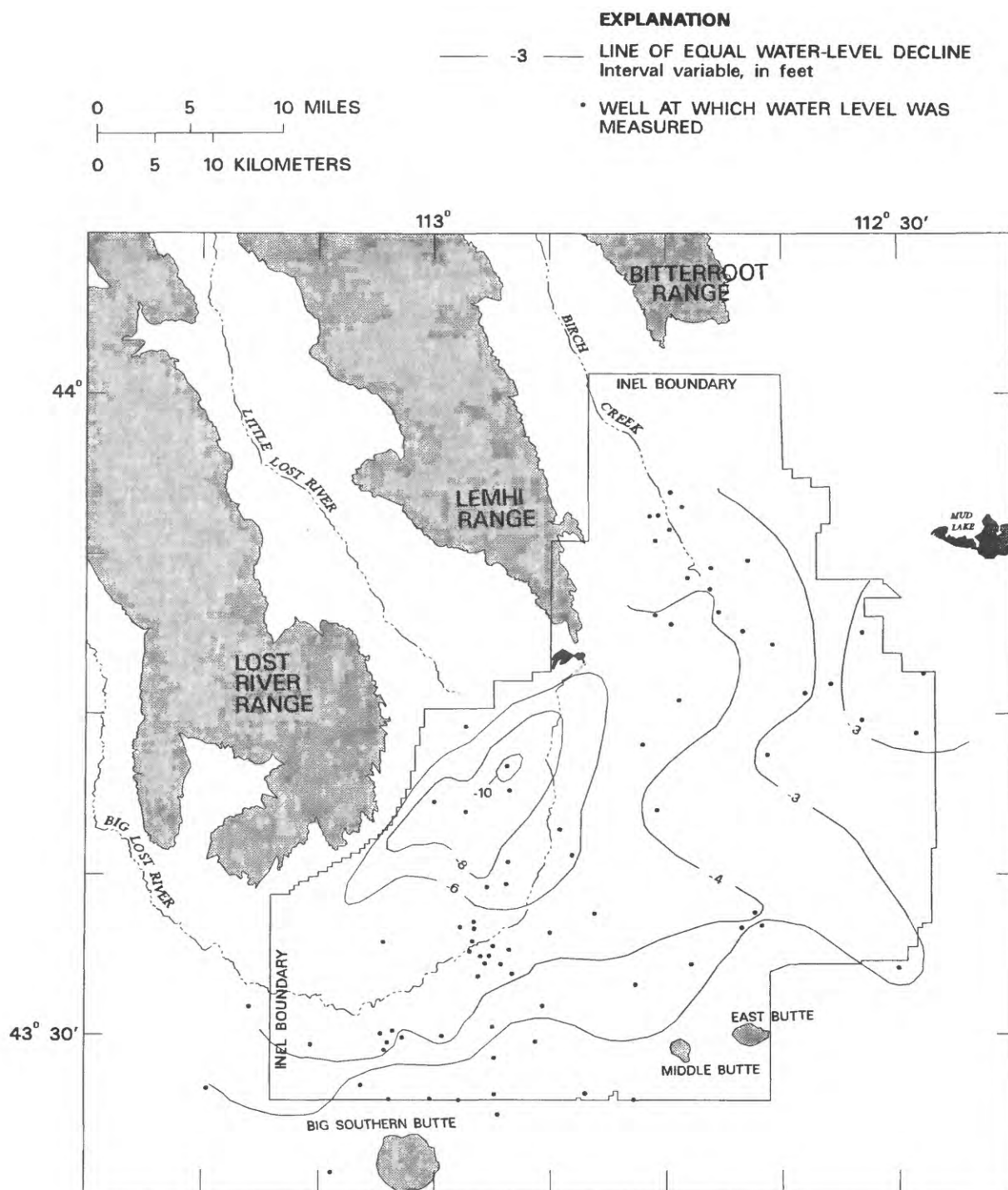


Figure 4. Generalized decline in ground-water levels in the Snake River Plain aquifer in the vicinity of the Idaho National Engineering Laboratory, April–August 1988 to April–August 1991.

Basaltic rocks form under conditions of rapid cooling and typically have a matrix that is finely crystalline or glassy. They tend to be enriched with minerals that crystallize slowly at high temperatures and great depths (Verhoogen and others, 1970, p. 281-282). For example, at atmospheric pressure, the melting temperatures of magnesium-rich olivine and pyroxene are about 1,890 and 1,557°C, respectively (Verhoogen and others, 1970, p. 268, table 6-1). When basaltic magmas erupt at the Earth's surface, they typically are in the temperature range of 1,000 to 1,200°C (Cas and Wright, 1987, p. 19, table 2.3). Rightmire and Lewis (1987b, p. 10) estimated that temperatures for liquid basalt flows at the RWMC were approximately 1,050°C. It follows that mineral crystals that formed at the higher temperatures and pressures at depth became trapped in the finely crystalline matrix that formed when the magma was extruded at the Earth's surface. Nace and others (1956, p. 99) observed this phenomenon in basalt at the INEL.

The Snake River Plain aquifer system is a series of extrusive basaltic lava flows and cinder beds that, in places, cover alluvium and lakebed sedimentary deposits. Individual flows range from 10 to 50 ft in thickness, although the average thickness may be from 20 to 25 ft (Mundorff and others, 1964, p. 143). The mineralogy of surficial material, sedimentary interbed samples, and basalt samples from the Snake River Plain aquifer system was determined by X-ray diffraction analysis (Bartholomay and others, 1989; Bartholomay and Knobel, 1989; Bartholomay, 1990) and by microscopic examination of basalt thin sections.

Surficial Material

* The alluvium and lakebed deposits that make up the surficial material of the eastern Snake River Plain contain a large component of loess or eolian dust (Rightmire and Lewis,

1987b, p. 20). Although anhydrite was not found in this study, Wood and Low (1988, p. D10) noted that it is present in eolian dust. Potential sources of anhydrite are the sedimentary rocks in tributary basins northeast and east of the eastern Snake River Plain (Robertson and others, 1974, p. 48, 50-51). Wood and Low (1988, p. D10) also noted that calcite is widely distributed in the surficial material.

A summary of bulk mineralogy for surficial sediment from the Big Lost River, Little Lost River, and Birch Creek drainages was presented by Bartholomay (1990, table 1). Quartz made up about 30 to 40 percent of the samples and was the most abundant mineral. Plagioclase feldspar made up about 10 to 20 percent of the samples, and clay minerals and detrital mica made up about 10 percent of channel deposits and 20 to 30 percent of non-channel deposits. The clay minerals identified in the samples were—in order of decreasing abundance—illite, smectite, mixed-layer clays, kaolinite, and chlorite (Bartholomay and others, 1989, p. 27; Bartholomay and Knobel, 1989, p. 9-12). Potassium feldspar made up about 10 percent of the samples, calcite made up about 5 to 25 percent, and pyroxene made up about 5 to 15 percent (except for Birch Creek channel deposits, which contained only trace amounts of pyroxene). Dolomite made up about 5 percent of the samples except those from the Big Lost River channel deposits, which were devoid of dolomite.

The mineralogy of surficial material at the RWMC that was described by Rightmire and Lewis (1987b, p. 20-21) is similar to that described by Bartholomay (1990, table 1). Nace and others (1956, p. 144-148) described the silt and clay mineralogy of selected surficial sediments from the INEL that also is similar to the description given by Bartholomay (1990). Nace and others (1956, p. 148) also observed that most clay minerals in the surficial deposits are detrital sediments.

Sedimentary Interbeds

The sedimentary interbeds are similar to the surficial deposits. They are made up of alluvium and lakebed deposits that contain a large component of eolian dust or loess (Nace and others, 1956, p. 183). Detrital anhydrite may be present in the eolian dust and caliche is widely distributed in the sedimentary layers (Wood and Low, 1988, p. D10). Fossil shells have been observed locally in interbed sediments (Nace and others, 1956, p. 184).

A summary of bulk mineralogy for sedimentary interbeds at the RWMC, Test Reactor Area (TRA), Idaho Chemical Processing Plant (ICPP), and TAN was presented by Bartholomay (1990, table 3). The mineralogical data used for that summary were taken from Barraclough and others (1976, table A-V, p. 123-124), Rightmire (1984, table 5, p. 17), Rightmire and Lewis (1987b, table 7, p. 35), Bartholomay and others (1989, table 11, p. 30), and Bartholomay (1990, table 2, p. 9). Quartz made up about 20 to 40 percent of the samples and was the most abundant mineral. Total feldspar made up about 10 to 30 percent of the samples. Thirteen samples of sedimentary-interbed material from wells near the ICPP, RWMC, and TRA contained 18 percent plagioclase and 13 percent potassium feldspar (Bartholomay and others, 1989, table 12, p. 31). Total clay minerals made up about 10 to 20 percent of the samples. The clay minerals identified in the samples were—in order of decreasing abundance—illite, smectite, mixed-layer clays, kaolinite, and chlorite (Bartholomay and others, 1989, table 13, p. 32; Bartholomay, 1990, table 2, p. 9). Detrital mica was not observed in the samples. Pyroxene made up about 10 to 20 percent of the samples, except at TAN, where it was not present. Dolomite and olivine were present in some samples. Calcite was the predominant mineral in interbed samples from TAN (Bartholomay, 1990, table 2, p. 9).

Rightmire (1984, p. 20-21) and Rightmire and Lewis (1987b, p. 28-33) observed that clay minerals—especially illite—are mostly detrital and that grains commonly have reddish coatings of ferric oxyhydroxides (FeOOH). Rightmire and Lewis (1987b, p. 36) observed the weathering of olivine to form FeOOH.

Basalt

The basalt flows that make up a large part of the material underlying the eastern Snake River Plain are remarkably uniform in mineralogical composition (Rightmire and Lewis, 1987b, p. 10; Lanphere and others, 1993, p. 4; Lanphere and others, 1994, p. 4). The differences in megascopic properties such as texture, color, and density are due largely to differences in emplacement and solidification (Nace and others, 1956, p. 97; Wood and Low, 1988, p. D7). Nace and others (1956, p. 97) concluded that the basalt flows were derived from a common magma reservoir; however, in a study of the subsurface at the INEL, Lanphere and others (1993, p. 8) concluded that more than one source of magma may have contributed to the accumulation of basalt flows.

The relative abundances of minerals in basalt samples from TAN Corehole-1 and the NPR Test well (fig. 1) were determined by X-ray diffraction for this study (tables 1-2). In addition, the mineralogy of five samples from TAN Corehole-1 was determined by a 500-point-count thin-section analysis (table 1). The minerals identified by X-ray diffraction analysis were—in order of decreasing abundance—plagioclase feldspar, pyroxene, olivine, ilmenite, hematite, and magnetite. The samples from TAN Corehole-1 at depths below land surface of 184, 187, and 236 ft had mineralogies consistent with those of the X-ray samples. The approximate abundances were plagioclase, 35 to 40 percent; pyroxene, 15 to 20 percent; olivine, 5 to 10 percent; combined ilmenite and magnetite, 0 to 10 percent; and hematite,

Table 1—Bulk mineralogy of basalt samples by X-ray diffraction analysis and thin-section analysis for TAN Corehole-1

[Bulk analyses (by X-ray diffraction): dom indicates mineral is dominant; maj indicates mineral is major in abundance; min indicates mineral is present in a minor amount; tr indicates mineral is present in a trace amount; poss indicates mineral is possibly present; ND indicates not detected. Percents from thin-section analyses determined by a 500-point count. Opaques: includes magnetite and ilmenite; NA indicates the determination is not appropriate for the type of analytical method. Thin sections were not made for samples from 204, 227, and 254 feet below land surface]

Interval sampled (feet below land surface)	Bulk analyses								
	Plagio- clase feldspar	Pyroxene	Olivine	Hematite	Opaques				
					Ilmenite (X-ray)	Total (thin section)	Magnetite (X-ray)	Glass matrix	Vesicles
184	38	20	9	2	NA	12	NA	9	11
187	35	14	7	5	NA	3	NA	11	26
204	dom	maj	maj	min	maj	NA	poss	ND	ND
227	dom	maj	maj	min	min	NA	poss	ND	ND
236	38	17	11	1	NA	7	NA	10	15
245	38	3	2	2	NA	ND	NA	32	23
245	dom	maj	min	min	min	NA	poss	ND	ND
254	dom	maj	maj	poss	ND	NA	poss	ND	ND
279	dom	maj	min	tr	min	NA	poss	ND	ND
279	31	2	6	2	NA	1	NA	45	14

Table 2—Bulk mineralogy of basalt samples by X-ray diffraction analysis for the NPR Test well

[Bulk analyses (by X-ray diffraction): dom indicates mineral is dominant; maj indicates mineral is major in abundance; min indicates mineral is present in a minor amount; tr indicates mineral is present in a trace amount; poss indicates mineral is possibly present; ND indicates not detected]

Interval sampled (feet below land surface)	Bulk analyses					
	Plagioclase feldspar	Pyroxene	Olivine	Hematite	Ilmenite	Magnetite
35	dom	maj	tr	tr	min	poss
121	dom	maj	min	ND	tr	ND
218	dom	maj	min	ND	min	ND
323	dom	maj	min	ND	ND	ND
435	dom	maj	maj	min	tr	ND
521	dom	dom	maj	min	ND	min

less than 5 percent. An additional 10 percent of the samples were finely crystalline matrix or glass and about 10 to 25 percent were vesicles. The thin-section samples from 245 and 279 ft below land surface contained less than 5 percent pyroxene and 30 to 45 percent matrix or glass. Other minerals were present in about the same proportions as the shallower samples. X-ray analyses of the samples from 245 and 279 ft indicated that pyroxene was the second most abundant mineral, which implies that pyroxene is present in the matrix in a cryptocrystalline form. Nace and others (1956, p. 100) observed a similar basalt mineralogy. One sample was about 80 percent finely crystalline groundmass and contained about 40 percent plagioclase and about 40 percent mixed pyroxene minerals. Wood and Low (1988, p. D7) also noted that the matrix generally has a mineralogy similar to the more coarsely crystalline part of the basalt. The larger phenocrysts were olivine and plagioclase and the smaller ones were olivine, labradorite (a form of plagioclase), clinopyroxene, ilmenite, magnetite, apatite, and glass. Lanphere and others (1993, p. 19) and Lanphere and others (1994, p. 22) used petrographic analysis to identify an identical mineral assemblage and a bimodal crystal-size distribution. Overall, the groundmass and glass made up about 20 percent of the rock and voids made up about 15 to 40 percent (Wood and Low, 1988, p. D7-D8). The molar composition of minerals in one basalt sample was as follows (Wood and Low, 1988, p. D8): olivine (0.43 magnesium and 0.57 iron); pyroxene (0.34 calcium, 0.39 magnesium, and 0.27 iron); and labradorite (0.5 calcium and 0.5 sodium).

Fractures and exposed vesicles near basalt-sediment interfaces are usually filled with detrital sediments (Wood and Low, 1988, p. D8). Particle orientation indicated that much of the fill material in RWMC samples was derived from water-borne sedimentation (Rightmire, 1984, p. 32). Secondary mineralization was observed by Wood and Low (1988,

p. D8) in vesicular basalt. Secondary chlorite, illite, smectite, calcite, and quartz were present in an outcrop sample.

The mineralogy of vesicle filling and fracture filling in basalt from TAN Corehole-1 was determined by X-ray diffraction analysis (table 3). The fill material was predominantly clay and quartz with some feldspar, calcite, and pyroxene. The clay minerals included illite, kaolinite, mixed layer, smectite, and chlorite. Illite was the most abundant clay mineral in the fracture and vesicle fillings.

Thin-section analyses of RWMC samples summarized by Rightmire (1984, p. 25-28, 32) indicated that minimal alteration of basalt minerals had occurred; however, chemical precipitates of silica and carbonate, and iron-oxyhydroxide rinds were sometimes present. The TAN Corehole-1 thin sections analyzed during this study consistently had iron stains coating the olivine phenocrysts. The iron coating—probably iron oxyhydroxide—made up about 2 percent of the samples. Rightmire and Lewis (1987a, p. 59-70) observed authigenic clay minerals, iron oxyhydroxides, calcite, silica, and possibly zeolites as weathering products or chemical precipitates that formed in place. Nace and others (1956, p. 94-98) also noted iron- and calcite-rich deposits associated with the basalt. Lanphere and others (1993, p. 20) noted that alteration of basalt minerals was minor in the samples they examined and was limited to weathering of olivine crystals and matrix glass.

General Considerations

The concentrations of naturally occurring inorganic constituents in ground water are controlled by the type, abundance, and reaction rates of minerals in the aquifer. The ability of water, especially water enriched with carbon dioxide (CO₂), to dissolve minerals was unequivocally demonstrated by experimentation in 1848 (Rogers and Rogers, 1848,

Table 3—Mineralogy of bulk and clay samples by X-ray diffraction analysis for vesicle filling and fracture filling in basalt from TAN Corehole-1

[Sample identifier: -I indicates vesicle filling in basalt; -F indicates fracture filling in basalt. Bulk analyses:----12----, number is the sum of percents of plagioclase and potassium feldspar. Clay analyses: dom indicates mineral is dominant; maj indicates mineral is major in abundance; min indicates mineral is present in a minor amount; tr indicates mineral is present in a trace amount; poss indicates mineral is possibly present; ND indicates not detected]

Sample identifier	Interval sampled (feet below land surface)	Bulk analyses (in percent mineral abundance)							
		Quartz	Plagioclase feldspar	Potassium feldspar	Calcite	Pyroxene	Olivine	Total clays	Hematite
TAN-CRH-F	184	14	----12----		74	0	0	0	0
TAN-CRH-I	187	36	19	9	0	17	0	19	0
TAN-CRH-F	204	8	----8----		0	0	0	84	0
TAN-CRH-I	227	7	----6----		0	0	0	87	0
TAN-CRH-F	236	8	----8----		0	0	0	83	0
TAN-CRH-F	245	15	----11----		6	0	0	68	0
TAN-CRH-F	279	12	----9----		10	0	0	69	0

Sample identifier	Interval sampled (feet below land surface)	Clay analyses (by abundance category)							
		Mixed layer	Smectite	Illite	Kaolinite	Chlorite	Other minerals		
							Quartz	Feldspar	Calcite
TAN-CRH-F	184	ND	ND	ND	ND	ND	min	min	dom
TAN-CRH-I	187	ND	ND	dom	ND	ND	min	tr	ND
TAN-CRH-F	204	poss	ND	min	dom	ND	ND	ND	ND
TAN-CRH-I	227	poss	poss	min	dom	ND	ND	ND	ND
TAN-CRH-F	236	dom	dom	maj	min	ND	ND	ND	ND
TAN-CRH-F	245	min	min	dom	poss	poss	tr	ND	ND
TAN-CRH-F	254	maj	maj	maj	poss	poss	ND	ND	ND

p. 401-405). Rogers and Rogers (1848) conducted tests on a variety of powdered rocks, minerals, organic compounds, and manmade materials and determined that up to 1 percent of the total mass was removed from the solid phase in as little as 48 hours. Additional early evidence establishing the solubility of minerals in water was summarized by Clarke (1924, p. 481-486). Nearly all minerals react with ground water to some extent; however, minerals that react slowly have less effect on the chemical composition of ground water than do minerals that react rapidly (Chapelle, 1983, p. 546). Goldich (1938) studied the weathering of igneous and metamorphic rocks and proposed a silicate mineral-stability series associated with rock weathering. He attributed the differences in mineral stability to the changing equilibrium conditions during formation of the minerals (such as changing temperature and pressure in a magma), which were described by Bowen (1922). Goldich (1938, p. 56) suggested that equilibrium conditions at the time of silicate-mineral formation are significantly different from surface conditions and that this is the reason for mineral weathering and the variability in rates of mineral weathering. For example, plagioclase is less stable under surface conditions than potassium feldspar, which in turn is less stable than crystalline quartz (Goldich, 1938, p. 56).

Garrels (1976, p. 76-78), on the basis of normative mineral calculations, inferred a “rank list” of minerals and their relative rates of reaction.” This list is in general agreement with Goldich’s (1938, p. 56) stability series. The stability of other minerals, such as calcite, is influenced more by changes in the concentration of dissolved CO₂ than by changes in ambient conditions. This is because the minerals formed at surface or near-surface conditions of temperature and pressure, and did not crystallize from a cooling magma.

The relative abundance of minerals in an aquifer also has an effect on water chemistry.

For example, Garrels (1976, p. 77) concluded that the rate of alteration of hornblende in a rhyolite is greater than that of plagioclase feldspar (on the basis of normative mineral calculations), even though the calculations indicated that plagioclase makes a larger contribution of ions to the reconstructed water chemistry. These observations and the fact that the predominant mineral in the rhyolite is plagioclase indicate that dissolution of minerals that constitute a large percentage of the parent material (but are less reactive) can contribute to water chemistry. Therefore, in this report, it is assumed that Snake River Plain aquifer water chemistry is dominated by the faster reacting, more abundant minerals and that the effects of slow-reacting minerals are negligible.

The order of crystallization of minerals in basalt from the Snake River Plain aquifer system was summarized by Nace and others (1956, p. 101). Magnetite crystallizes first, followed by olivine, plagioclase, and simultaneous crystallization of titanite and ilmenite. On the basis of the findings of Goldich (1938), Nace (1956), and Garrels (1976), the stability of minerals in the Snake River Plain aquifer system basalts, in order of increasing stability, is magnetite, olivine, plagioclase, titanite and ilmenite. Titanite is a titanium-rich clinopyroxene mineral. Although magnetite and ilmenite are unstable minerals, the abundances are small relative to olivine, plagioclase, and pyroxene. As a result, magnetite and ilmenite do not impact Snake River Plain aquifer water chemistry as much as the other minerals.

Ion Distribution

The distribution of major ions in ground water is partially controlled by the solubilities of minerals in the aquifer and by the ground-water flow system. Concentrations of solutes in ground water generally increase in the direction of ground-water flow until equilib-

rium between the solid, liquid, and gaseous phases is established. Once equilibrium is established, concentrations remain relatively constant until the equilibrium of the system is disrupted by other factors such as microbial activity, industrial or agricultural waste disposal, a change in mineralogy, or mixing with water from another source. A comparison of mean concentrations along the generalized direction of ground-water flow in the eastern Snake River Plain aquifer (fig. 5), for calcium, magnesium, sodium, potassium, bicarbonate, chloride, sulfate, and silica is shown in table 4. The report by Knobel and others (1992) summarizes chemical data from selected sites at or near the INEL. The column with sample size of 22 to 24 presents the mean concentration of each constituent for all samples regardless of proximity to potential waste-disposal areas. This data grouping reflects the influence of waste disposal at the INEL on natural water

chemistry. The column with sample size of 5 presents the mean concentration of each constituent for samples far enough away from waste-disposal sites to minimize influence from those sites. This data grouping more closely represents natural water chemistry. The report by Wood and Low (1988) summarizes chemical data from sites throughout the eastern Snake River Plain aquifer. The mean concentrations should be larger than the mean concentrations for natural water at the INEL because most samples summarized by Wood and Low (1988) were downgradient of the INEL. The reports by Bartholomay and others (1992, 1993, 1994a) summarize data from sites exclusively downgradient from the INEL.

The smallest mean concentrations of the constituents in table 4 are generally from sites at the INEL not affected by waste disposal (Knobel and others, 1992, n = 5). The mean

Table 4—Comparison of mean concentrations of selected constituents in the generalized direction of ground-water flow, eastern Snake River Plain aquifer, Idaho

[Authors: complete bibliographic citations are given in the section of this report on "selected references": Wood and Low (1988, table 9); Bartholomay and others (1992, tables 2 and 11; 1993, tables 2 and 13; 1994a, tables 2 and 12); Knobel and others (1992, tables 3 and 5-6). Abbreviation: n =, sample size equals. Units are milligrams per liter]

Constituent	Report			
	Bartholomay and others (1992, 1993, 1994a) n = 59	Wood and Low (1988) n = 424	Knobel and others (1992) n = 22 to 24	Knobel and others (1992) n = 5
Calcium	50	51	43	40
Magnesium	20	18	16	15
Sodium	33	26	21	12
Potassium	4.6	4.0	3.3	2.6
Bicarbonate	216	220	168	177
Chloride	38	28	35	14
Sulfate	48	41	30	21
Silica	34	31	28	29

concentrations of samples from all INEL sites are similar to mean concentrations of samples from the unaffected sites except for sodium, chloride, and sulfate. These constituents typically are associated with liquid wastes disposed of at the INEL

The mean concentrations of constituents in 424 samples from sites in the entire eastern Snake River Plain (table 4) were larger than the mean concentrations associated with the INEL samples not affected by waste disposal ($n = 5$). The increases in mean concentrations in these samples probably reflect the inclusion of more downgradient samples in the statistical analysis. The mean concentrations of the constituents from the 59 samples collected exclusively downgradient of the INEL (Bartholomay and others, 1992, 1993, 1994a) were similar to or larger than the mean concentrations of the 424 samples reported by Wood and Low (1988). The increases in mean concentrations of chemical constituents mimic the generalized configuration of the flow system in the eastern Snake River Plain aquifer (fig. 5).

Hydrochemical Facies

Hydrochemical facies are useful tools for describing the chemical character of ground water. The format for the hydrochemical facies diagram used in this report was developed by Piper (1944) and is similar to one designed by Hill (1940). The diagram allows the plotting of relative concentrations of major anions and cations and the chemical character of multiple water samples. Freeze and Cherry (1979, p. 249-250) and Hem (1985, p. 178-179) briefly described the diagram and its uses. Generally, only bicarbonate, chloride, and sulfate are considered when analyzing the hydrochemical facies of anions in ground water; however, data in Knobel and others (1992, table 8) indicate that nitrate is a significant constituent in Snake River Plain aquifer water. As a result,

nitrate is included with sulfate as a component of the anion hydrochemical facies.

The hydrochemical facies of water from selected wells and springs on or near the INEL, from wells at TAN, and from wells at the RWMC are shown on figures 6-8, respectively. The data for calculating the cation and anion facies and water character were taken from Knobel and others (1992, tables 5, 6, 8; sampling locations are shown on their figs. 2-4)

Figure 6 indicates that water from wells and springs in the southern half of the INEL and south of the INEL boundary (Knobel and others, 1992, fig. 2) generally are calcium-magnesium bicarbonate in character. This water character is typical of most of the water samples considered in this report.

Webb Spring discharges from rhyolitic rocks that form Big Southern Butte (fig. 1) and has an anomalous water chemistry. It is enriched with magnesium and sodium plus potassium and depleted with calcium relative to Snake River Plain aquifer samples (fig. 6). In addition, Webb Spring has a smaller dissolved solids content and alkalinity and a larger dissolved silica concentration (Knobel and others, 1992, tables 3, 5). These differences in water chemistry reflect a shorter water-residence time and the mineralogical differences between the rhyolitic rocks of Big Southern Butte and the basaltic rocks of the Snake River Plain aquifer.

The water sample from NRF-2 (fig. 6) was enriched with chloride and depleted with bicarbonate relative to the other water samples. In addition, the specific conductance was larger than the other water samples. These differences are likely associated with waste-disposal activities at the NRF (fig. 1).

Water samples from wells at TAN (fig. 7) showed an increase in the relative concentrations of sodium plus potassium and chloride and a corresponding decrease in calcium and

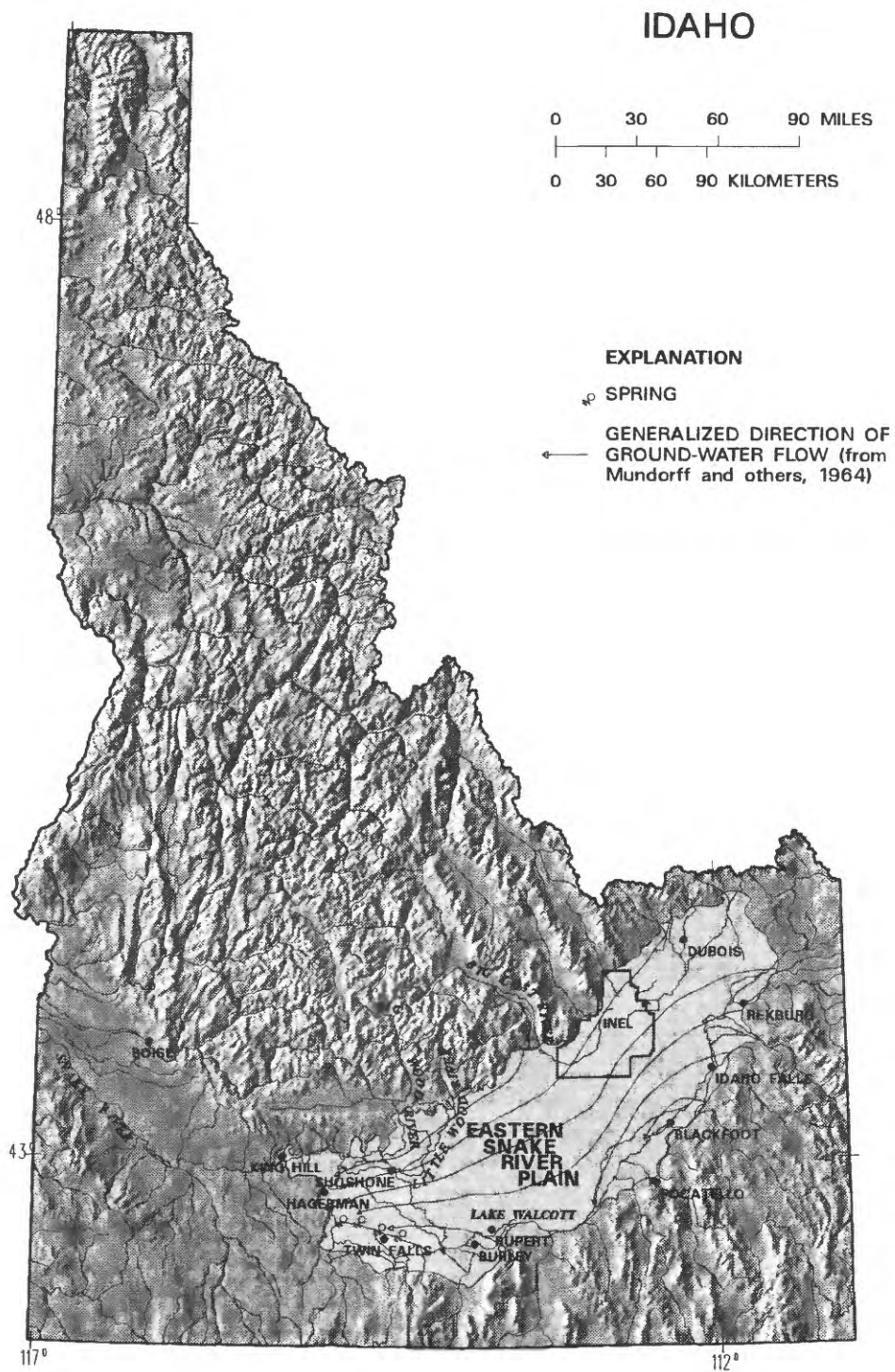


Figure 5. Generalized direction of ground-water flow in the eastern Snake River Plain aquifer and locations of the eastern Snake River Plain and the Idaho National Engineering Laboratory.

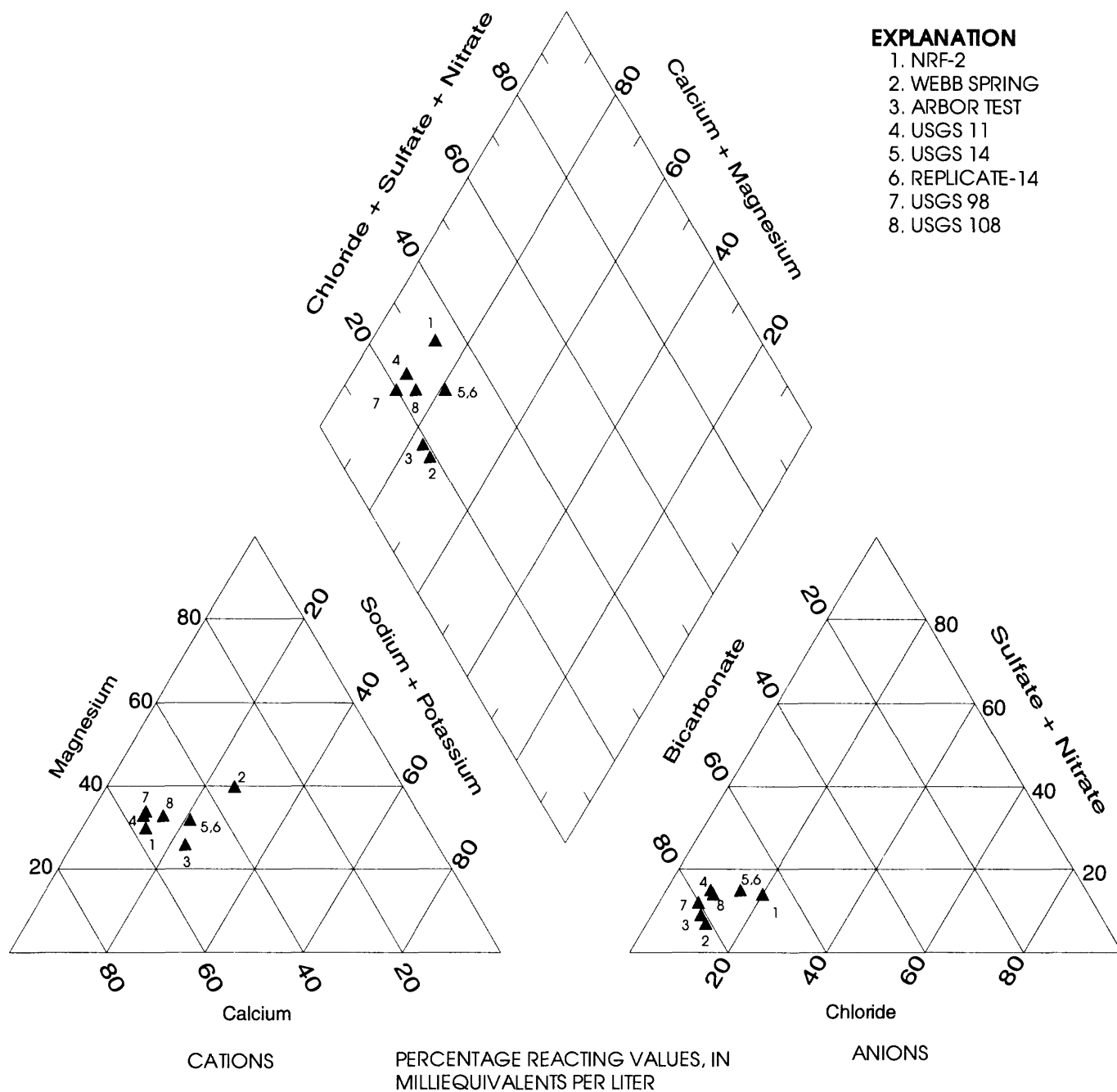


Figure 6. Major-ion composition of water from selected wells and Webb Spring on or near the Idaho National Engineering Laboratory, Idaho.

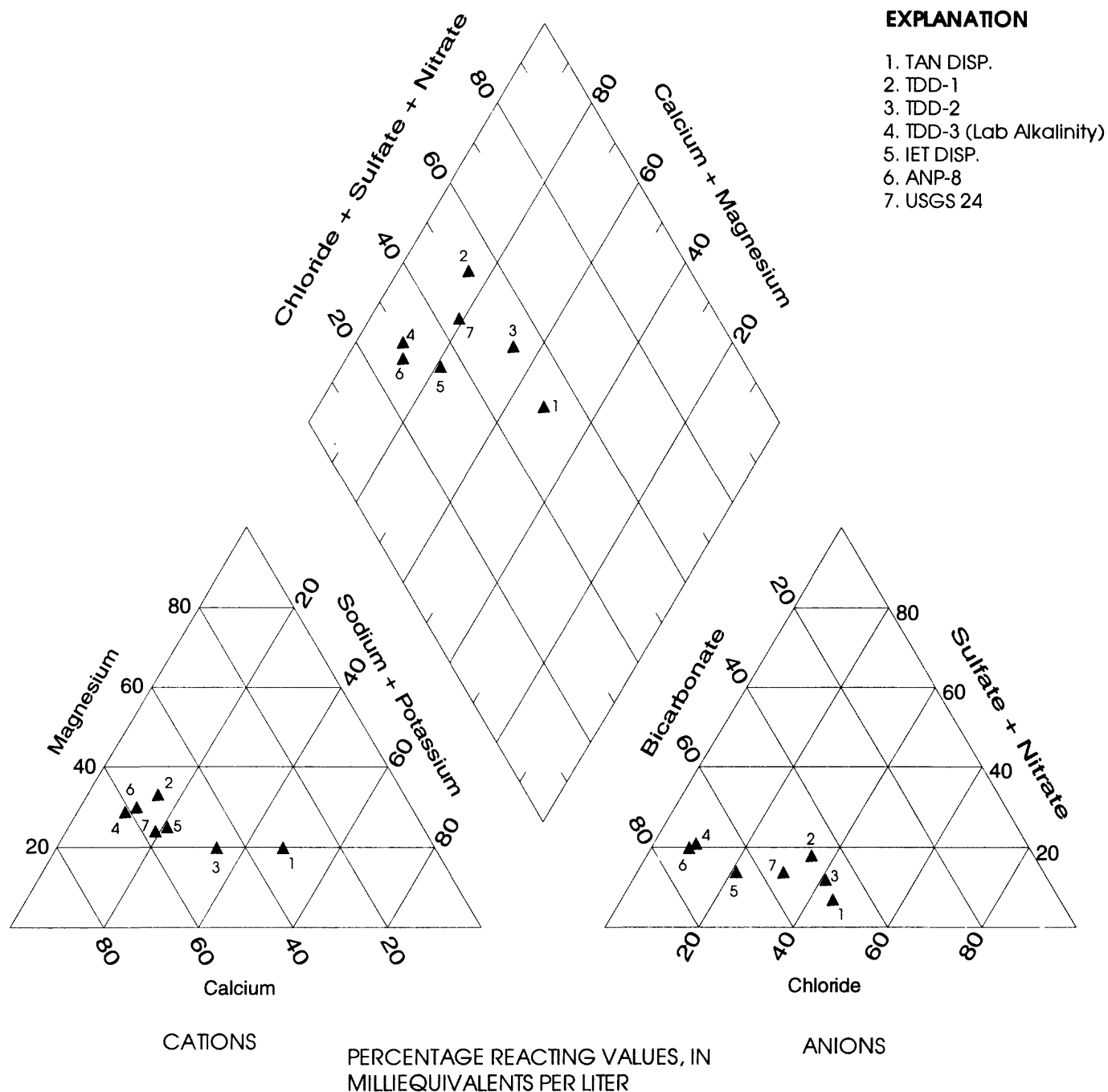


Figure 7. Major-ion composition of water from wells at the Test Area North, Idaho National Engineering Laboratory, Idaho.

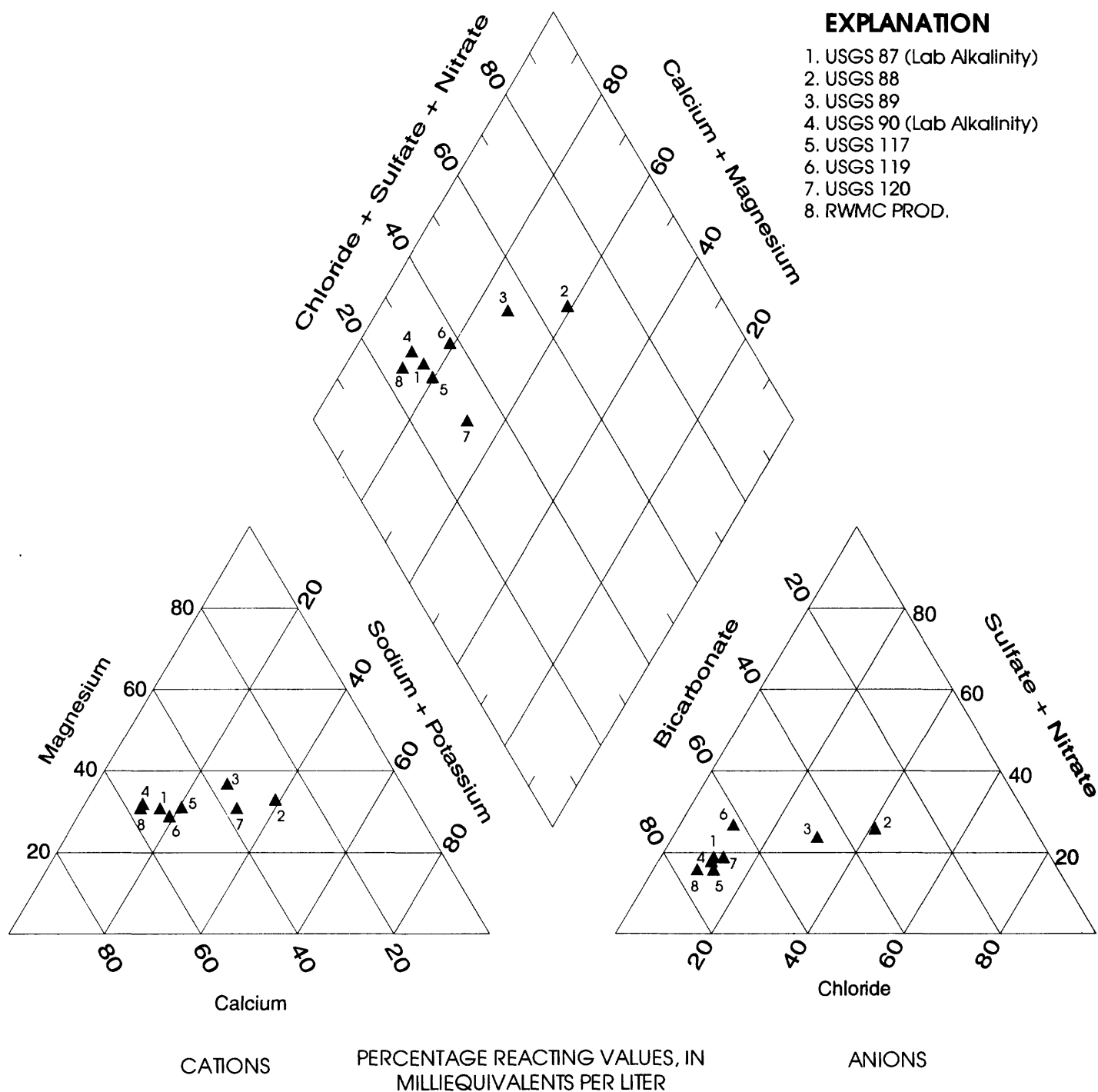


Figure 8. Major-ion composition of water from wells at the Radioactive Waste Management Complex, Idaho National Engineering Laboratory, Idaho.

bicarbonate when compared with other water samples considered in this report. The effect was most pronounced at the TAN Disposal well and generally decreased with distance from the well (Knobel and others, 1992, fig. 3). The differences in water chemistry at TAN are attributed to the injection of aqueous waste into the aquifer at the TAN Disposal well.

The hydrochemical facies of water samples from wells at the RWMC (fig. 8) were generally similar to those unaffected by waste disposal (fig. 6). Water samples from wells 88, 89, and 120, however, were slightly enriched with sodium plus potassium; those from wells 88 and 89 also were enriched with chloride.

Thermodynamic Considerations

Chemical equilibrium is attained when a chemical system, under constant pressure and temperature conditions, is at its smallest possible energy level. This occurs when the free energy of the system is zero. Chemical systems tend to give off energy by means of chemical reaction until equilibrium is achieved.

The deviation of a system from equilibrium can be expressed by determining the saturation index (SI) of a water that has a given chemical composition with respect to a mineral. SI is defined as follows:

$$SI = \log \frac{IAP}{K}, \quad (1)$$

where IAP = ion activity product,
and

K = the equilibrium constant for the reaction.

An SI of zero indicates that the water is in equilibrium with respect to a reaction. A negative SI indicates that the water is undersaturated, and a positive SI indicates that the water is supersaturated. Saturation indices for analyses of the 22 samples reported by Knobel and others (1992, tables 5, 6) that were considered in this report were determined by

the computer program WATEQF (Plummer and others, 1978).

Saturation indices for analyses of water samples with respect to selected minerals that are present in the Snake River Plain aquifer or that might potentially be precipitated from aqueous solution are shown for carbonate, sulfate, and silicate minerals on figures 9-11, respectively. Maximum, minimum, and median saturation indices were plotted for each of several minerals for each of three data groupings: (1) wells at TAN, (2) wells at the RWMC, and (3) selected wells on or near the INEL. Uncertainties associated with the calculation of saturation indices largely result from uncertainties inherent in the thermodynamic constant—free energy of formation (ΔG_f)—of the various solid and dissolved phases. The solid phases shown on figures 9-11 have ΔG_f values that are well established and should not add significant uncertainty to the calculated saturation indices. The variability of the ranges of data shown on figures 9-11 resulted from two principal sources: (1) natural or induced variations in water chemistry at the multiple sample points, and (2) any analytical error associated with the water-sample analyses.

The median SI values for the carbonate minerals aragonite, calcite, and dolomite (fig. 9) indicate that the Snake River Plain aquifer water samples were supersaturated with respect to these minerals; therefore, precipitation of the minerals is the only thermodynamic possibility. Conversely, median SI values with respect to the carbonate minerals magnesite, strontionite, and witherite (fig. 9) indicate undersaturation; therefore, dissolution of these minerals is the only thermodynamic possibility. The median SI values with respect to fluorite (fig. 10), the sulfate minerals anhydrite, celestite, and gypsum (fig. 10), and the silicate minerals diopside, clinoenstatite, and olivine (fig. 11) indicate undersaturation and, hence, dissolution of these minerals if

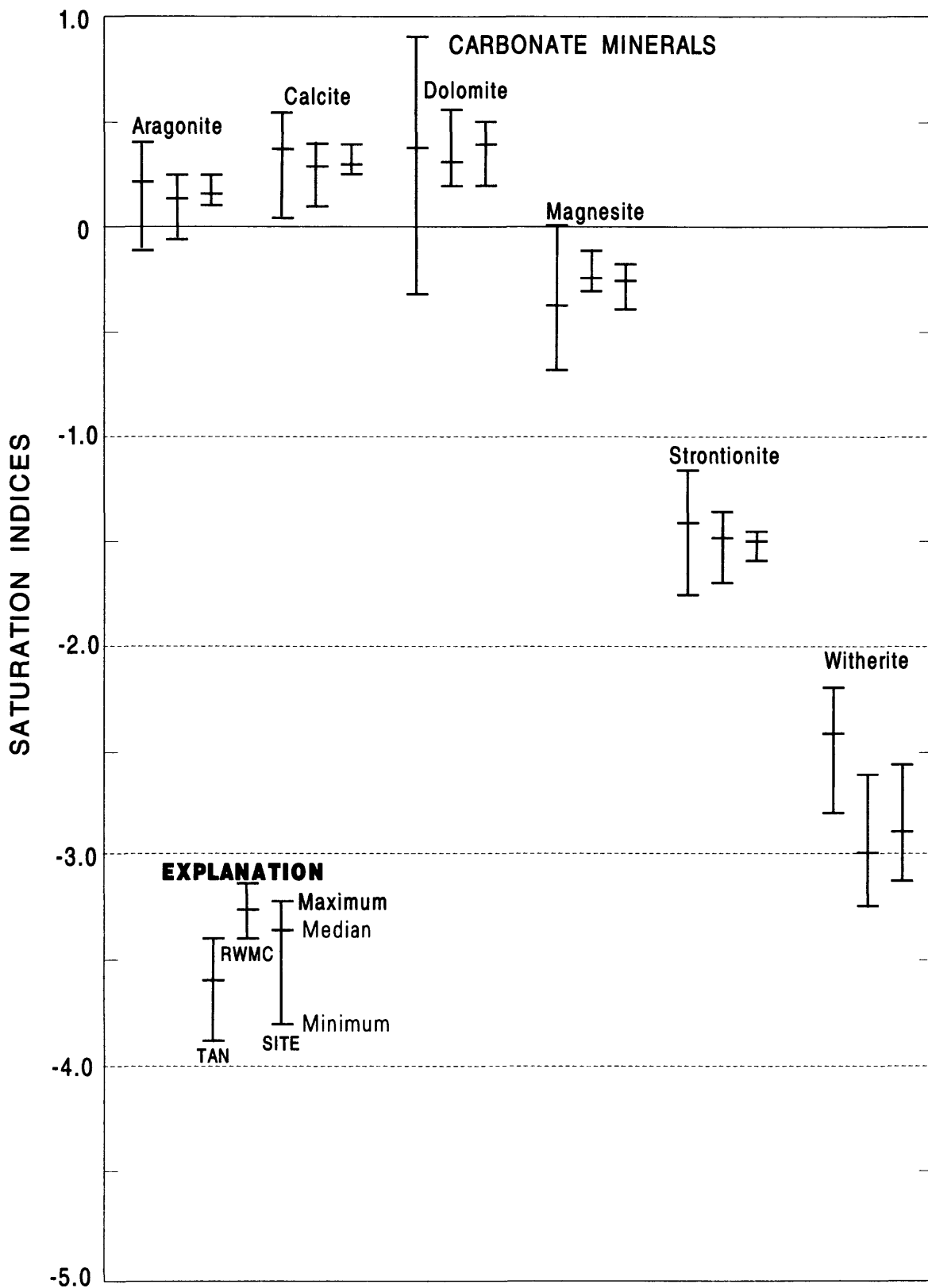


Figure 9. Saturation indices for Snake River Plain aquifer water samples from seven wells at TAN, eight wells at the RWMC, and seven selected wells at sites on or near the Idaho National Engineering Laboratory, Idaho with respect to selected carbonate minerals.

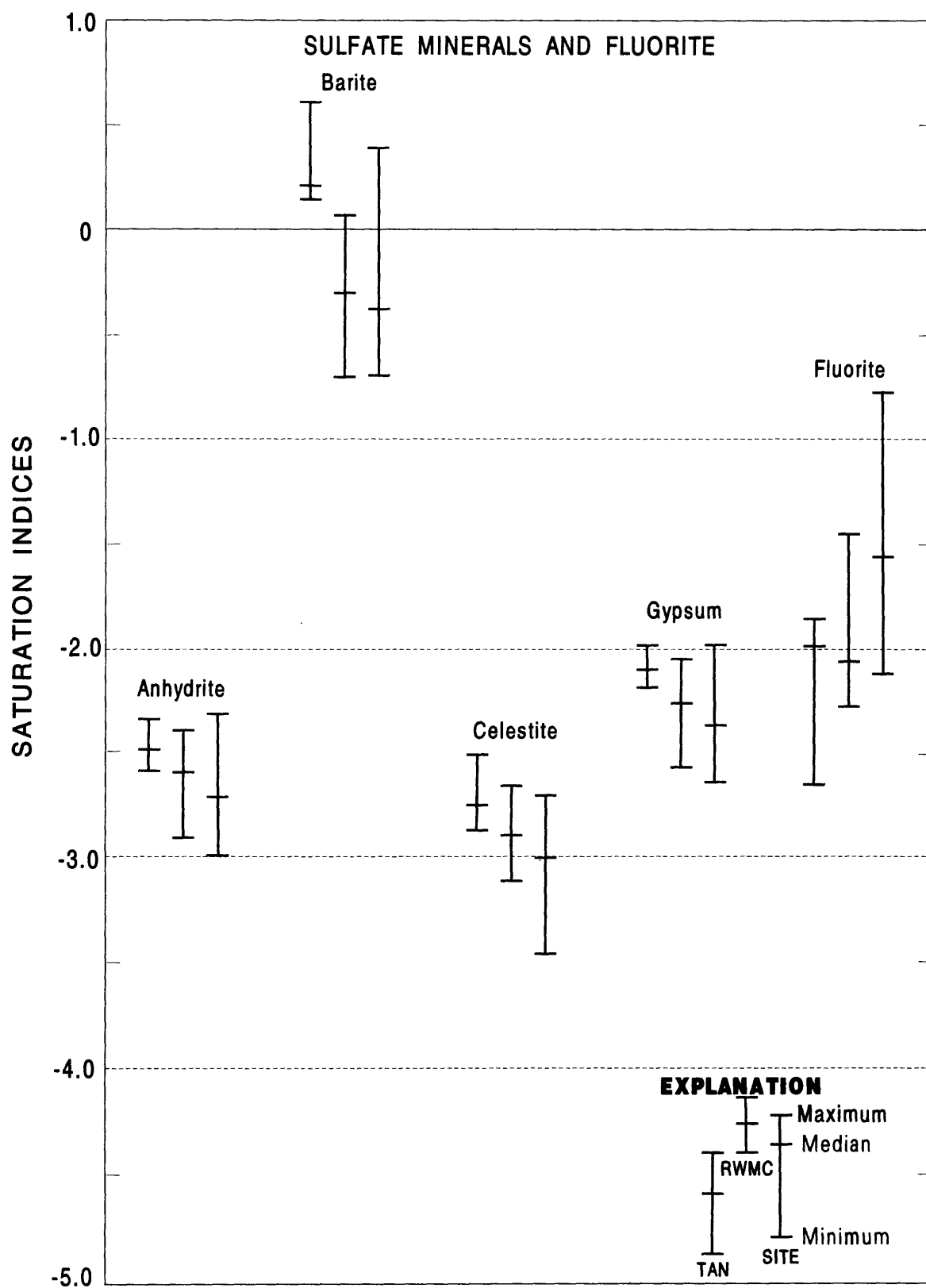


Figure 10. Saturation indices for Snake River Plain aquifer water samples from seven wells at TAN, eight wells at the RWMC, and seven selected wells at sites on or near the Idaho National Engineering Laboratory, Idaho with respect to selected sulfate minerals and fluorite.

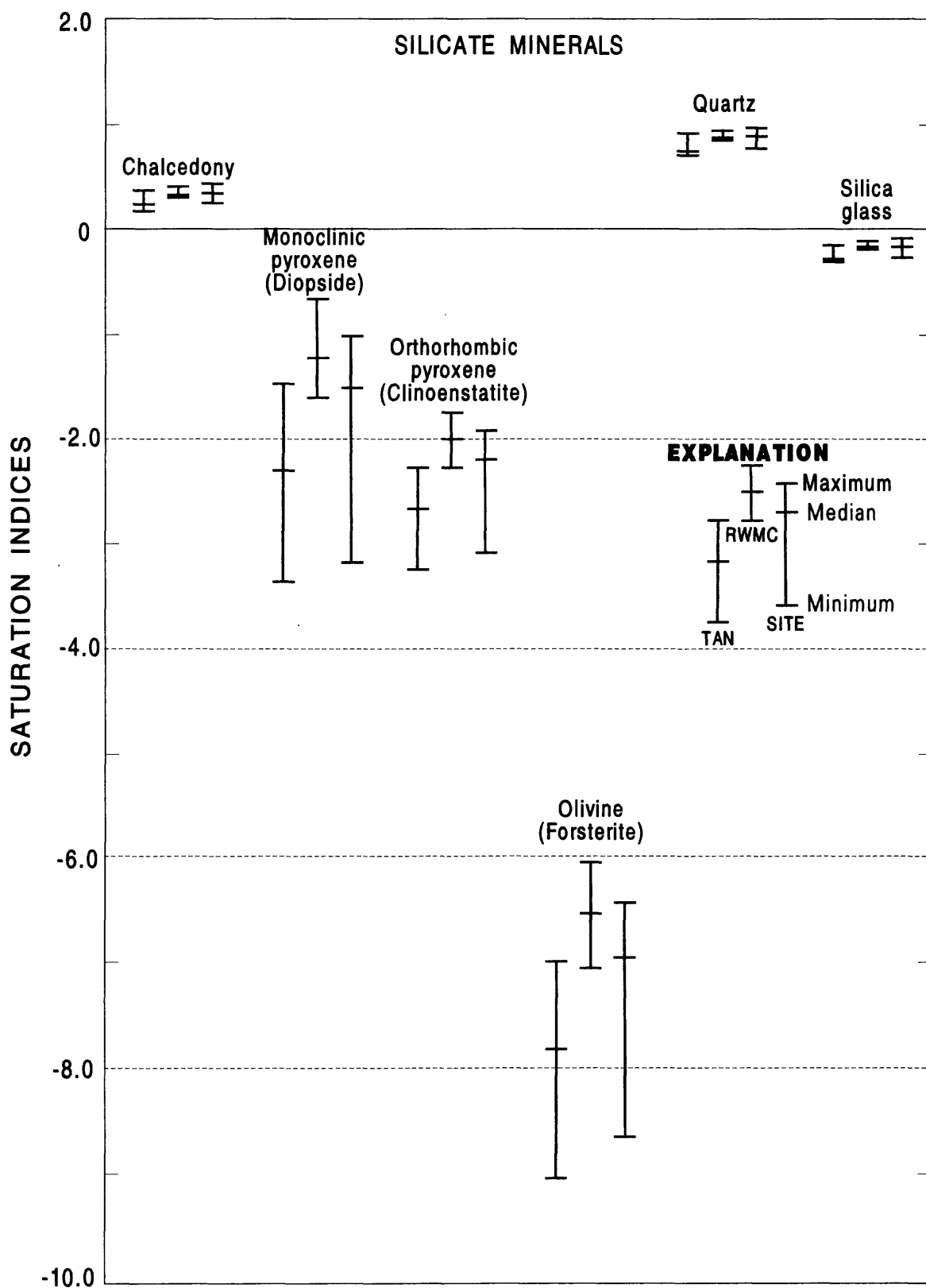


Figure 11. Saturation indices for Snake River Plain aquifer water samples from seven wells at TAN, eight wells at the RWMC, and seven selected wells at sites on or near the Idaho National Engineering Laboratory, Idaho with respect to selected silicate minerals.

present in the aquifer. The sulfate mineral barite (fig. 10) is thermodynamically stable in association with water from wells near TAN but unstable to metastable with respect to water from the other wells sampled. Median SI values with respect to crystalline quartz and cryptocrystalline chalcedony (fig. 11) indicate that they are stable in the solid phase. The median SI values with respect to amorphous silica glass (fig. 11) indicate that it is unstable in the solid phase; however, it is near equilibrium and the lack of stability is less certain.

The minerals discussed above dissolve congruently and the calculated SI values are reliable because they are based on well-known thermodynamic data. Complex aluminosilicate minerals, such as the feldspar minerals, dissolve incongruently and form residual clay minerals. The solid-phase thermodynamic data for such reactions are not well known and calculated SI values are less reliable predictors of mineral stability. In addition, dissolved aluminum concentrations are required to calculate SI values, and aluminum is difficult to measure in water samples because of its chemical affinity for colloidal material and the small concentrations at which it exists. In order to avoid these difficulties, the stability of aluminosilicate minerals commonly is evaluated by plotting water chemistry data on diagrams depicting the stability fields of the solid phases in a given chemical system. For example, in the system microcline-muscovite-gibbsite-kaolinite, $\log_{10} [K^+]/[H^+]$ is plotted as a function of $\log_{10} [Si(OH)_4]$ (the brackets indicate ion activity). This technique avoids the analytical problems associated with measuring aluminum concentrations. Water-chemistry data for samples from wells at TAN, wells at RWMC, and selected wells and a spring on or near the INEL were superposed on stability diagrams (figs. 12-14) for the potassium system. All the data plot in the kaolinite stability field, which indicates that microcline, muscovite, and gibbsite are unstable solid phases and would react with water to form kaolinite.

In a similar fashion, $\log_{10} [Ca^{+2}]/[H^+]^2$ is plotted as a function of $\log_{10} [Si(OH)_4]$ in figures 15-17 and $\log_{10} [Na^+]/[H^+]$ is plotted as a function of $\log_{10} [Si(OH)_4]$ in figures 18-20. Figures 15-17 indicate that for water associated with anorthite, calcium montmorillonite is the stable solid phase. Similarly, figures 18-20 indicate that for water associated with albite, kaolinite and sodium montmorillonite are the stable solid phases.

The actual chemical composition of plagioclase in the Snake River Plain aquifer system lies between the end-member compositions for albite and anorthite; hence, the phase boundaries involving plagioclase in figures 15-20 should be considered approximate. A similar uncertainty in the phase boundaries involving potassium minerals is effected in figures 12-14. Because of the uncertainties associated with aluminosilicate mineral stability diagrams, conclusions about water equilibrium and mineral stability that are based solely on the diagrams should be considered tentative. A detailed discussion of sources of uncertainty in mineral-stability diagrams is provided by Drever (1988, p. 113-114).

Plausible Chemical Reactions

Plausible chemical reactions are defined in this report as those likely to take place naturally in the Snake River Plain aquifer system because (1) the requisite source minerals, liquids, and gases are present in the system; (2) thermodynamic conditions allow the reaction to proceed; and (3) the dissolved-, gaseous-, or solid-product phases either are present in or can be accounted for by processes that remove them from the system.

Congruent Dissolution Reactions

Olivine.—Olivine is a common mineral in the basaltic rocks of the Snake River Plain aquifer system and also is present in some

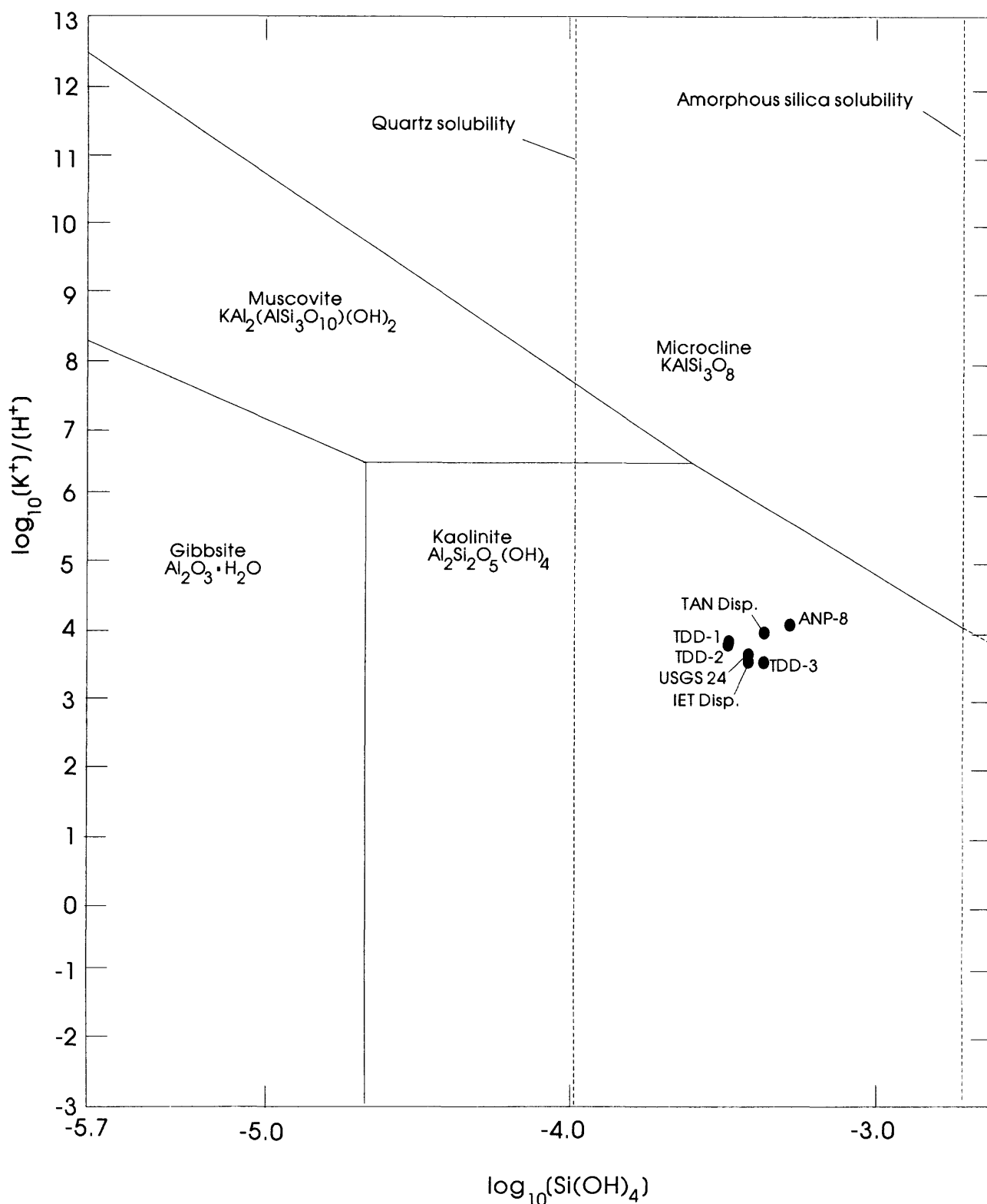


Figure 12. Stability relations among microcline, muscovite, gibbsite, and kaolinite with superposed compositions of water samples from the Snake River Plain aquifer near Test Area North. Modified from Freeze and Cherry (1979, p. 272). Brackets indicate thermodynamic activity of indicated species.

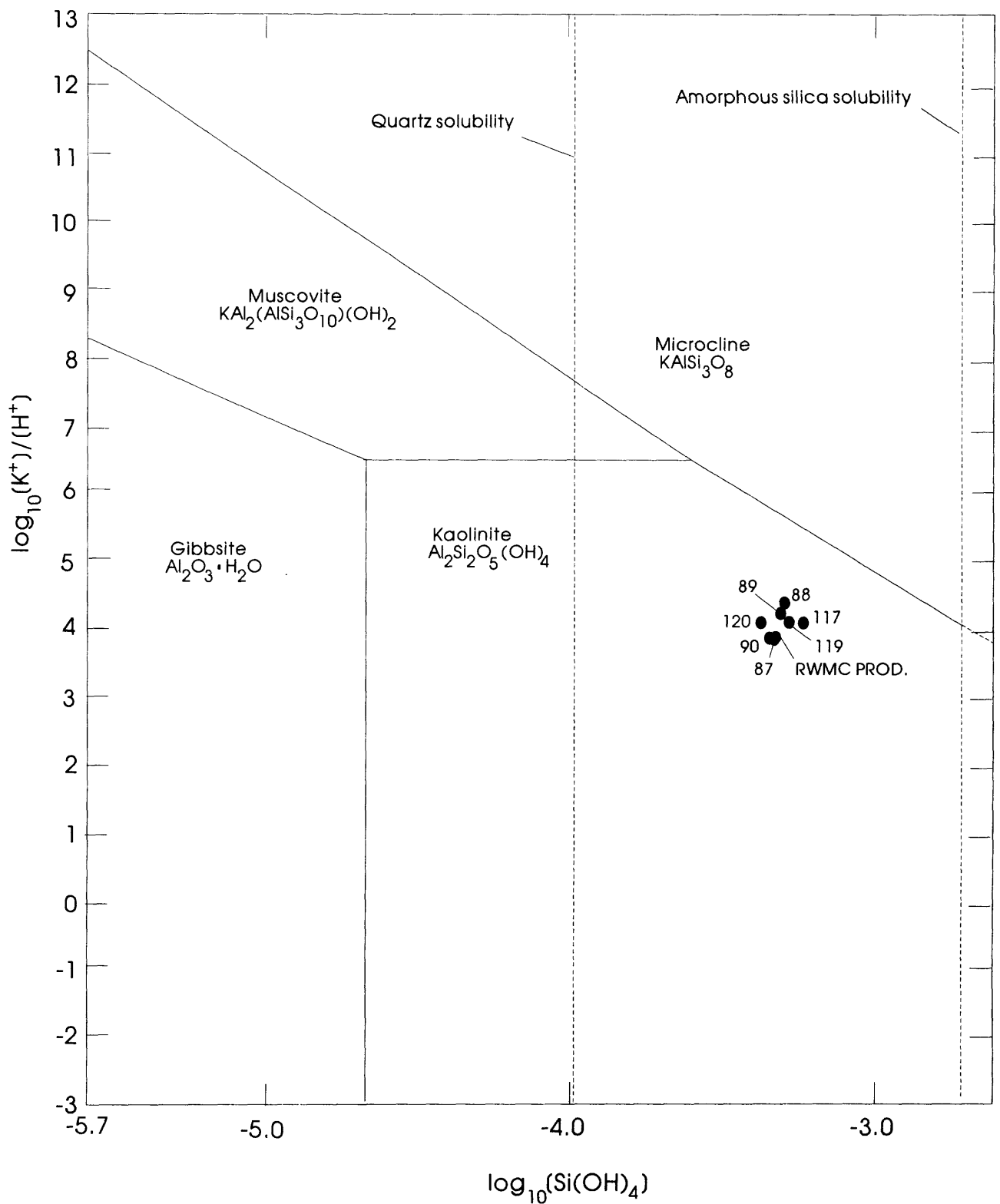


Figure 13. Stability relations among microcline, muscovite, gibbsite, and kaolinite with superposed compositions of water samples from the Snake River plain aquifer near the Radioactive Waste Management Complex. Modified from Freeze and Cherry (1979, p. 272). Brackets indicate thermodynamic activity of indicated species.

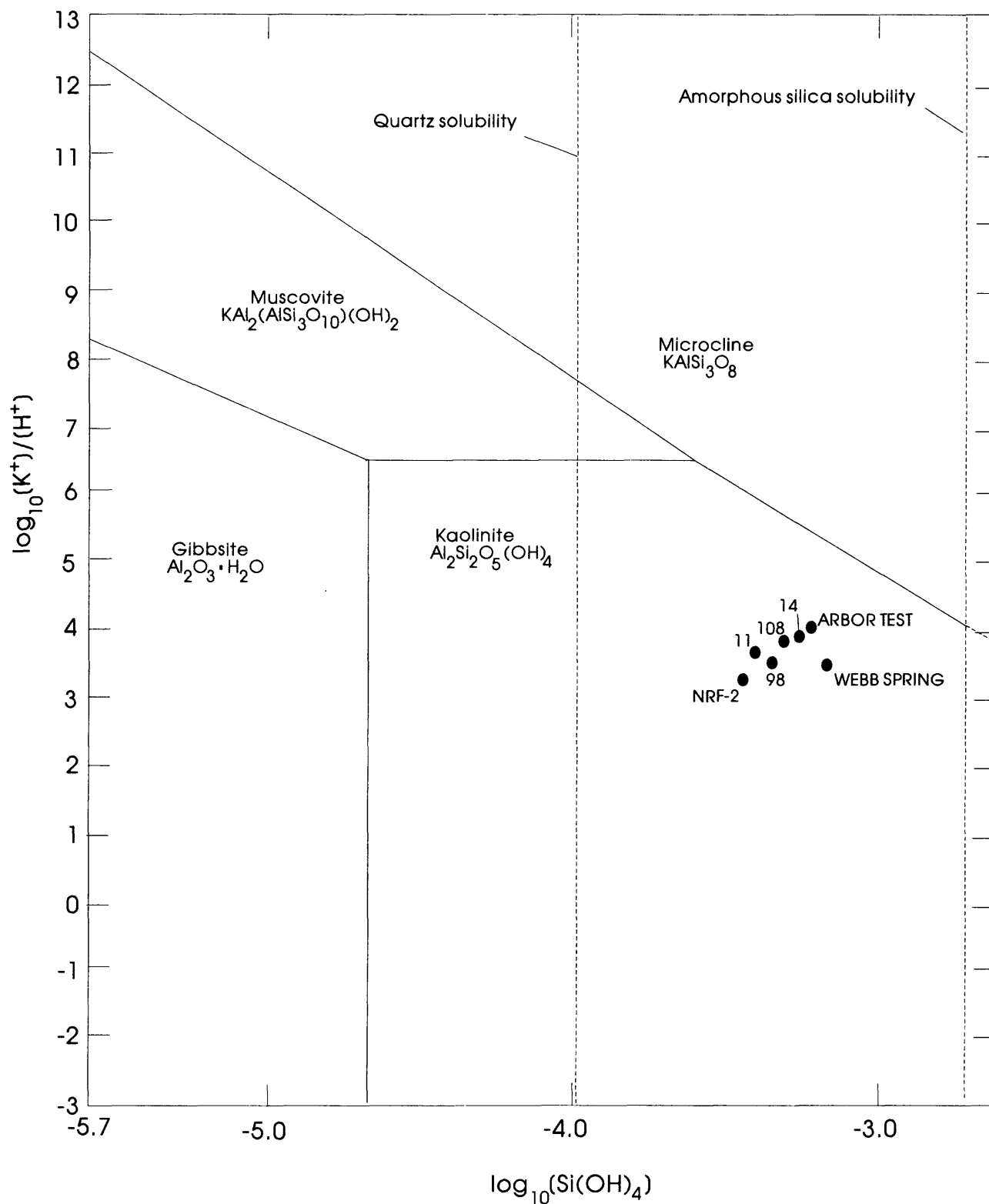


Figure 14. Stability relations among microcline, muscovite, gibbsite, and kaolinite with superposed compositions of water samples from selected wells completed in the Snake River Plain aquifer on or near the Idaho National Engineering Laboratory, and from Webb Spring. Modified from Freeze and Cherry (1979, p. 272). Brackets indicate thermodynamic activity of indicated species.

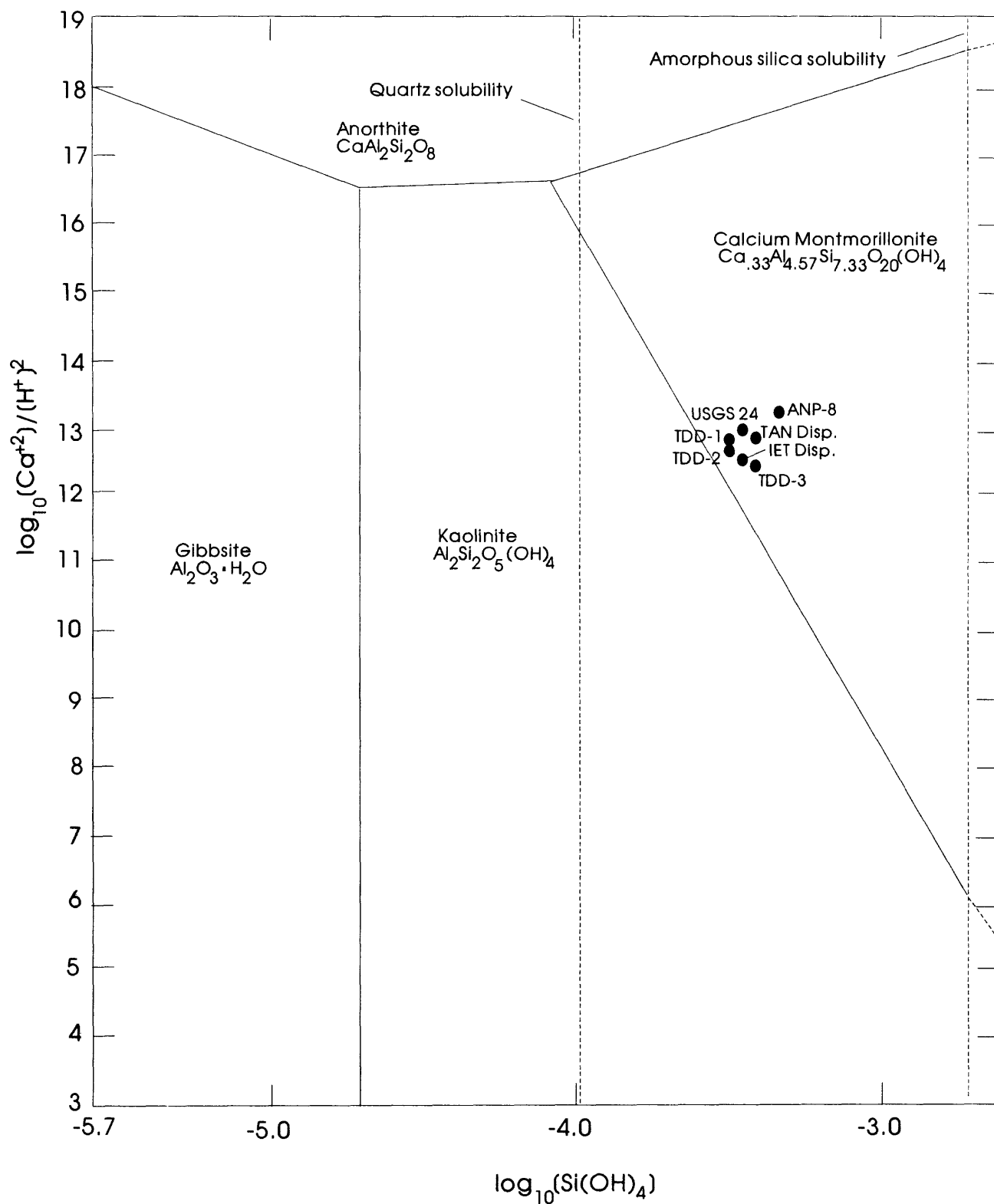


Figure 15. Stability relations among anorthite, gibbsite, kaolinite, and calcium montmorillonite with superposed compositions of water samples from the Snake River Plain aquifer near Test Area North. Modified from Freeze and Cherry (1979, p. 272). Brackets indicate thermodynamic activity of indicated species.

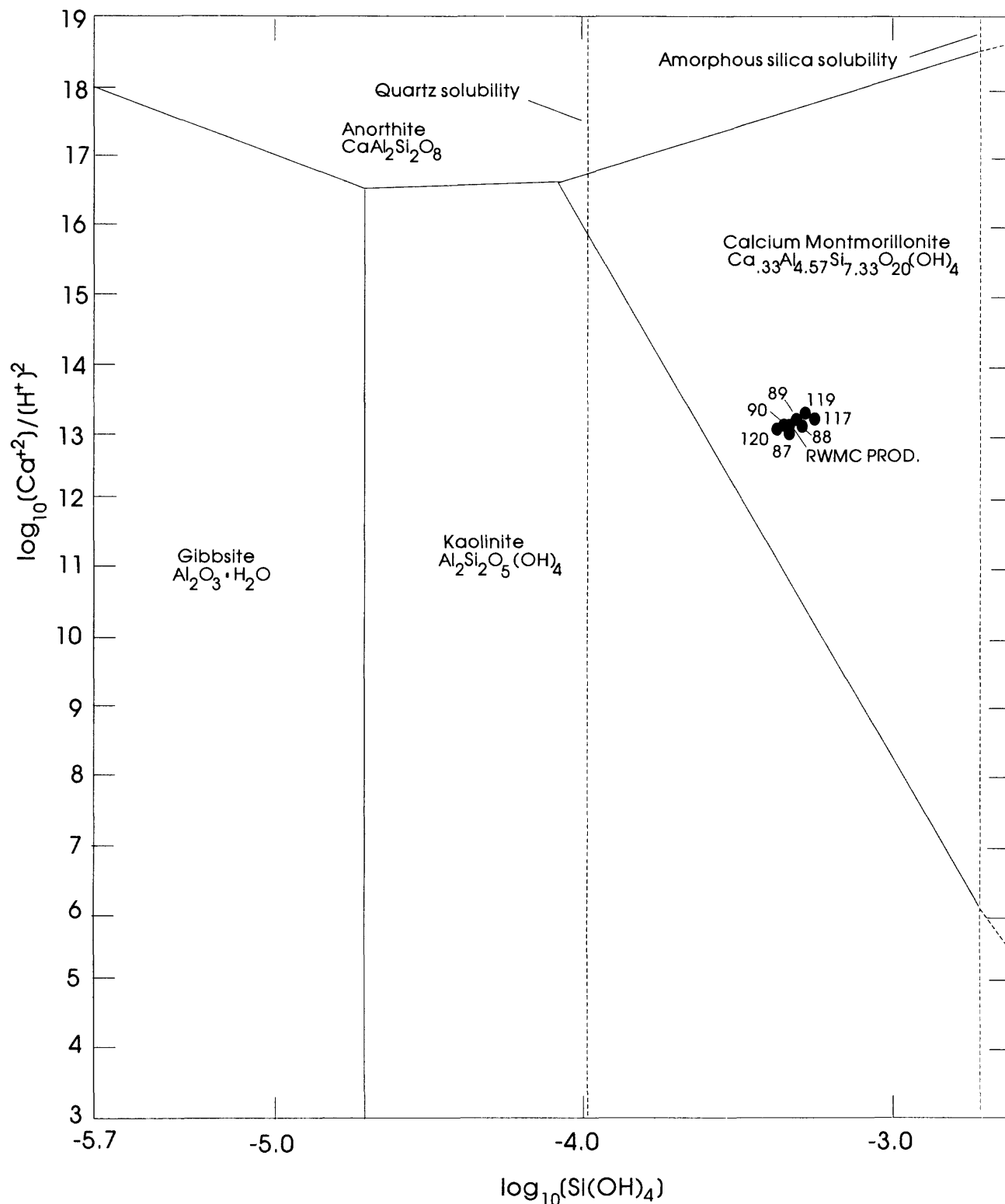


Figure 16. Stability relations among anorthite, gibbsite, kaolinite, and calcium montmorillonite with superposed compositions of water samples from the Snake River Plain aquifer near the Radioactive Waste Management Complex. Modified from Freeze and Cherry (1979, p. 272). Brackets indicate thermodynamic activity of indicated species.

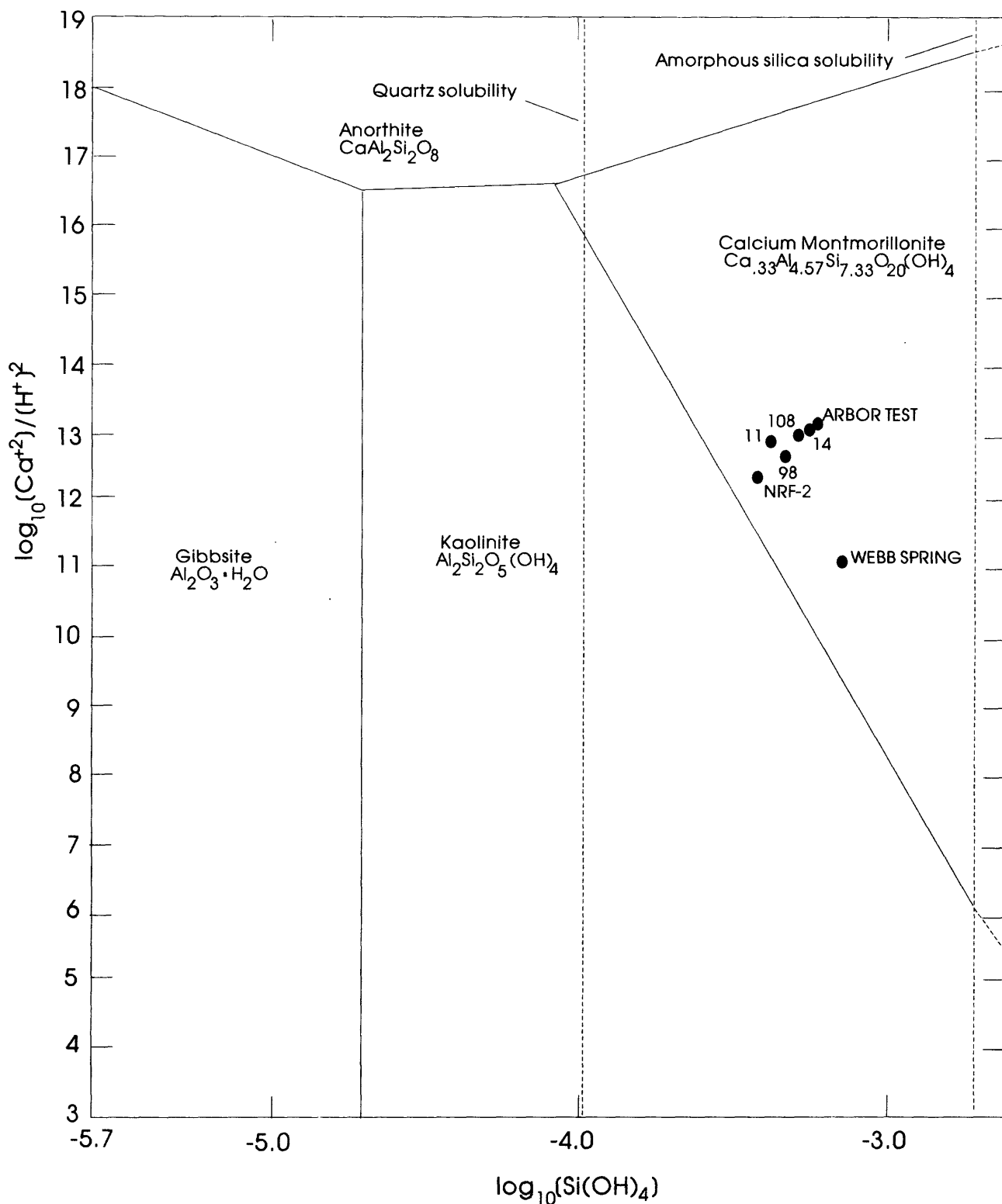


Figure 17. Stability relations among anorthite, gibbsite, kaolinite, and calcium montmorillonite with superposed compositions of water samples from selected wells completed in the Snake River Plain aquifer on or near the Idaho National Engineering Laboratory, and from Webb Spring. Modified from Freeze and Cherry (1979, p. 272). Brackets indicate thermodynamic activity of indicated species.

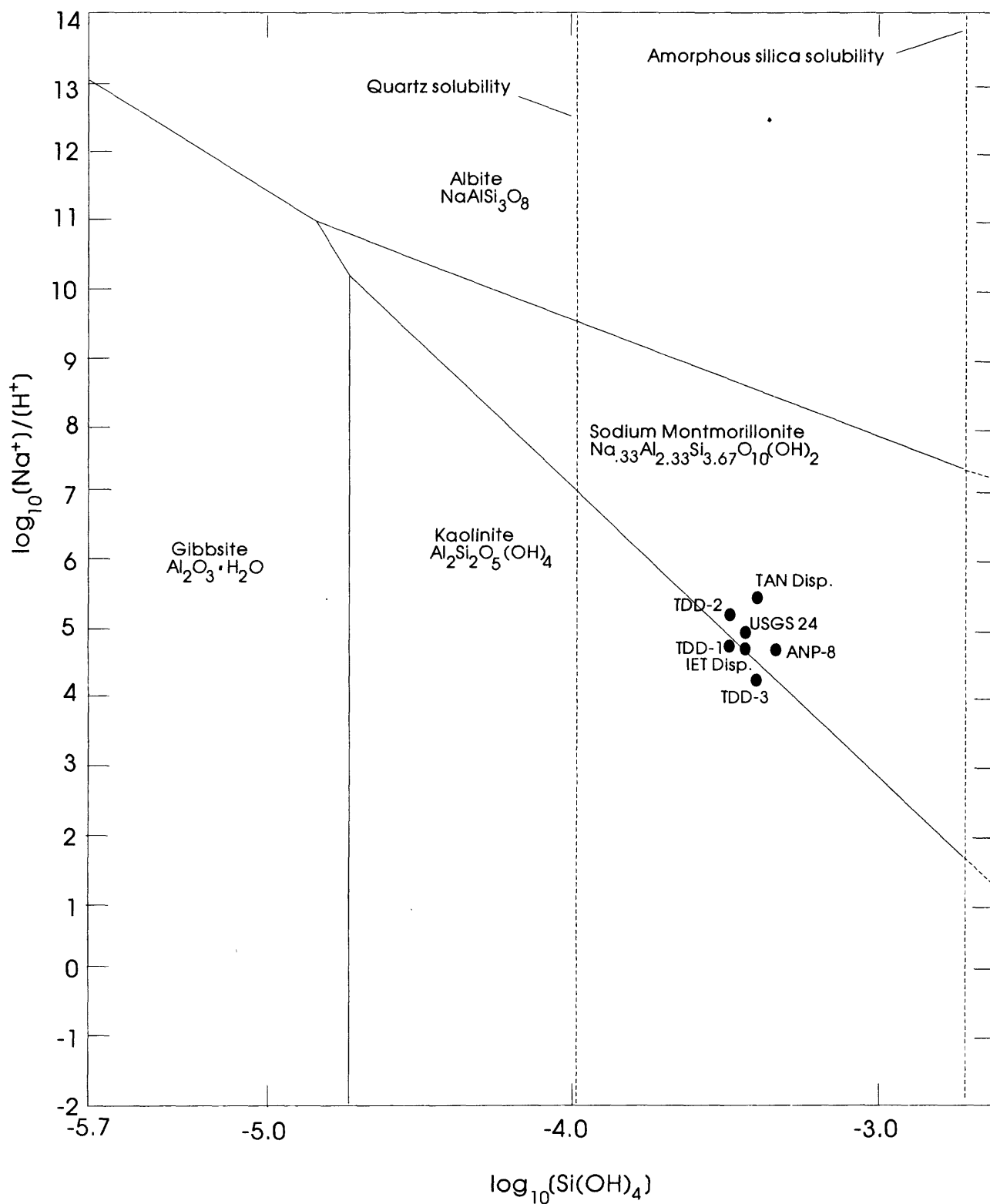


Figure 18. Stability relations among albite, gibbsite, kaolinite, and sodium montmorillonite with superposed compositions of water samples from the Snake River Plain aquifer near Test Area North. Modified from Freeze and Cherry (1979, p. 272). Brackets indicate thermodynamic activity of indicated species.

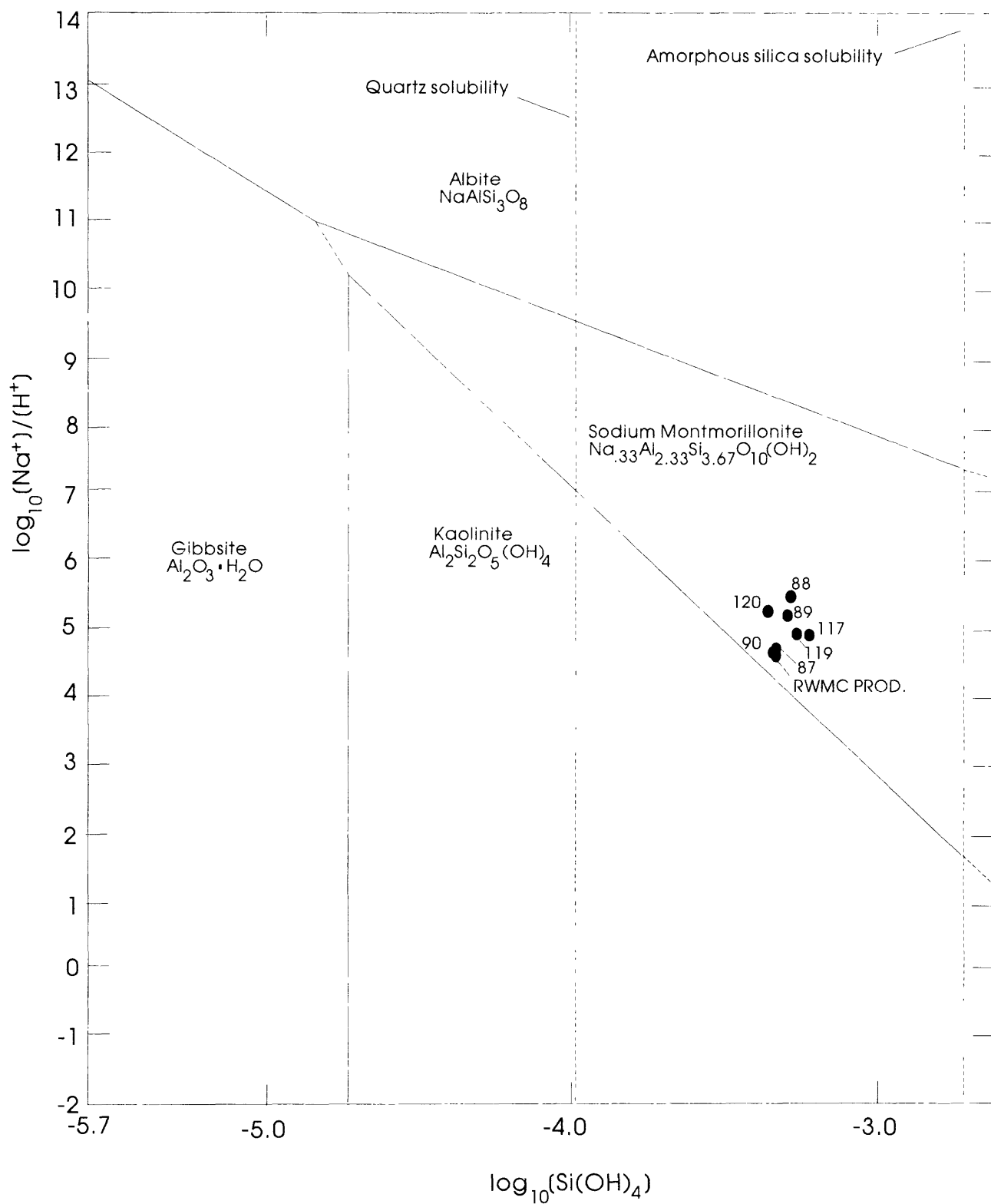


Figure 19. Stability relations among albite, gibbsite, kaolinite, and sodium montmorillonite with superposed compositions of water samples from the Snake River Plain aquifer near the Radioactive Waste Management Complex. Modified from Freeze and Cherry (1979, p. 272). Brackets indicate thermodynamic activity of indicated species.

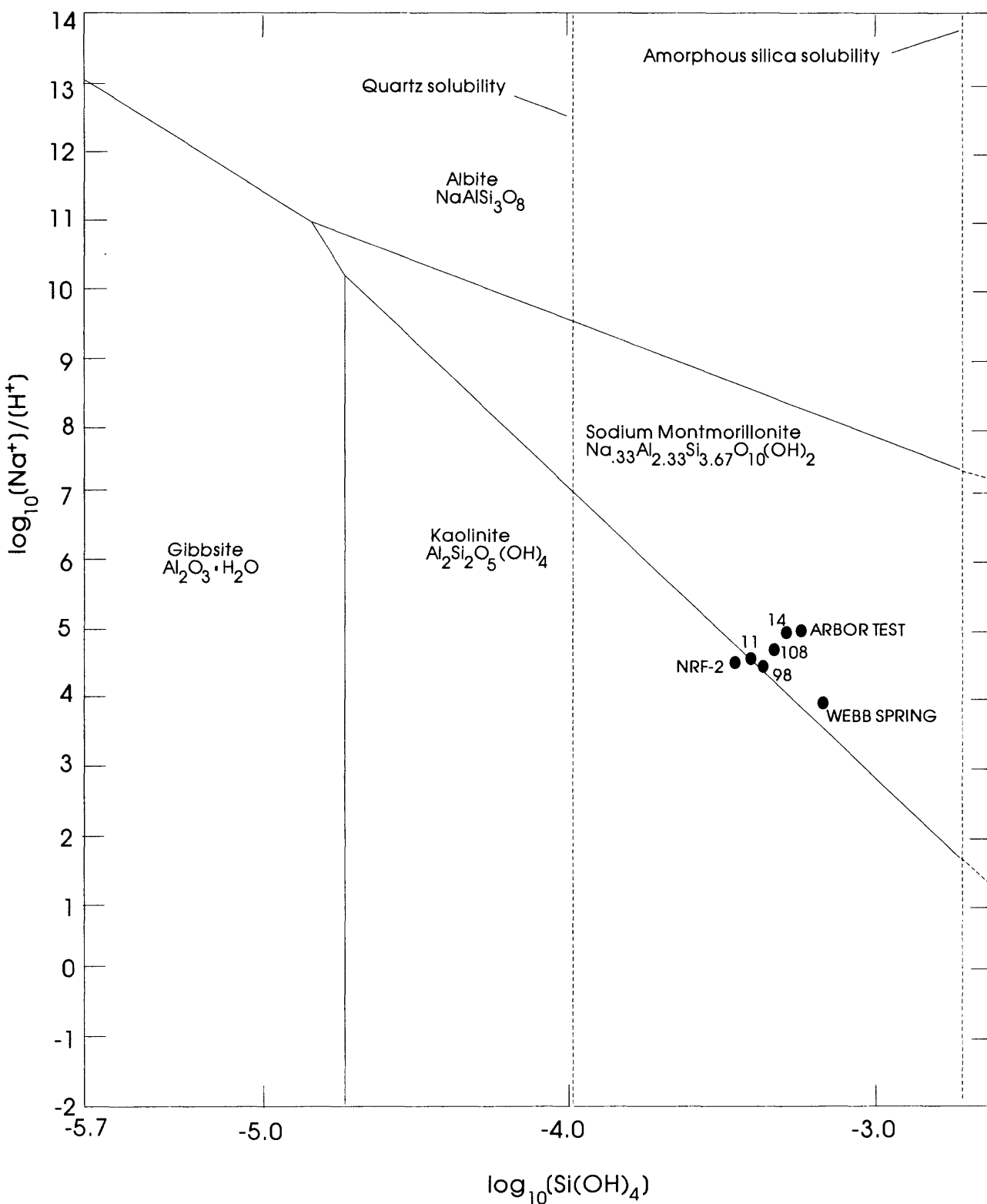


Figure 20. Stability relations among albite, gibbsite, kaolinite, and sodium montmorillonite with superposed compositions of water samples from selected wells completed in the Snake River Plain aquifer on or near the Idaho National Engineering Laboratory, and from Webb Spring. Modified from Freeze and Cherry (1979, p. 272). Brackets indicate thermodynamic activity of indicated species.

sedimentary interbeds. Mineral stability and thermodynamic considerations previously discussed in this report indicate that olivine is an extremely unstable mineral when exposed to Snake River Plain aquifer water. Wood and Low (1988, p. D8) reported an olivine composition of $(\text{Mg}_{0.43}\text{Fe}_{0.57})_2\text{SiO}_4$ for a single sample of olivine from basalt from the Snake River Plain aquifer system. Because the actual mineralogical composition of olivine probably differs from place to place, an idealized formula was substituted to simplify the reaction. Olivine in the presence of dissolved CO_2 , dissolved oxygen, and water reacts to form dissolved constituents and ferric hydroxide or one of its amorphous precursory phases such as ferric oxyhydroxide. When olivine dissolves, the ferrous form of iron in olivine is oxidized to ferric iron.

The association of olivine and ferric oxyhydroxide (Rightmire and Lewis, 1987b, p. 36) and/or iron stains in the form of rinds (Rightmire, 1984) are strong evidence that olivine in the aquifer is actively weathering. A chemical-reaction model for the weathering of olivine (equation 2) is given in table 5 at the end of this report.

Pyroxene minerals.—Pyroxene minerals make up a significant part (up to 20 percent) of Snake River Plain aquifer system materials. Pyroxene minerals have a wide range of chemical composition and structural form, and more than one type is present in the Snake River Plain aquifer system. Pyroxene minerals are present as macroscopic crystals and cryptocrystalline groundmass in basalt and as detrital grains in sedimentary interbeds and surficial material. Saturation indices calculated for water samples from the aquifer (fig. 11) indicate undersaturation with respect to monoclinic pyroxene (diopside) and orthorhombic pyroxene (clinoenstatite). Either mineral (along with pyroxene minerals of intermediate compositions and structures) should dissolve in association with aquifer water. The abun-

dance of pyroxene in the aquifer system suggests that dissolution of the mineral impacts water chemistry. Wood and Low (1988, p. D8) reported a composition of $(\text{Ca}_{0.34}\text{Mg}_{0.39}\text{Fe}_{0.27})\text{Si}_2\text{O}_6$ in a pyroxene crystal in a basalt core sample. Because of known differences in the chemical composition and structural form of pyroxene minerals in the Snake River Plain aquifer system, the simpler form of diopside was used to represent the pyroxene minerals in this report. Diopside dissolves in the presence of CO_2 and water to form dissolved constituents. If a more complex pyroxene mineral containing ferrous iron weathers, an oxidation process similar to the olivine reaction takes place. A chemical-reaction model for the dissolution of diopside (equation 3) is given in table 5. For comparison purposes, Wood and Low (1988, p. D20, equation 5) presented a chemical reaction model for the dissolution of an iron-bearing pyroxene mineral.

Anhydrite.—Anhydrite may be present as an accessory mineral in sedimentary interbeds and surficial deposits of the Snake River Plain aquifer system. The saturation indices of water samples with respect to anhydrite (fig. 10) indicate that it is an unstable mineral and dissolves to form dissolved calcium and sulfate. The contribution of anhydrite to the dissolved load of Snake River Plain aquifer water is constrained by its limited abundance in the aquifer system; however, locally the reaction may affect water chemistry. A chemical-reaction model for the dissolution of anhydrite (equation 4) is provided in table 5.

Incongruent Dissolution of Feldspar

Plagioclase feldspar is ubiquitous in the surficial material, sedimentary interbeds, and basalt flows that make up the Snake River Plain aquifer system. An estimated 10 to 50 percent of the matrix is plagioclase feldspar. Conversely, potassium feldspar is found only in the unconsolidated materials and makes up

about 10 percent of the bulk sediment composition. In addition, plagioclase generally is less stable than potassium feldspar. These observations suggest that plagioclase contributes more to the dissolved load of ground water than potassium feldspar. As a result, incongruent dissolution of plagioclase feldspar is considered to be the dominant feldspar reaction in this report.

The water-chemistry data plotted on figures 15-20 suggest that the plagioclase minerals anorthite and albite would weather to form dissolved constituents and clay minerals. Incongruent dissolution of anorthite would produce calcium montmorillonite and incongruent dissolution of albite would produce either sodium montmorillonite or kaolinite. These conclusions are tentative because of the uncertainties inherent in figures 15-20; however, incongruent dissolution of plagioclase feldspar in this aquifer appears possible. The hydrochemical environmental conditions identified by Rightmire and Lewis (1987b, p. 37) that are favorable for the formation of smectite are present in the Snake River Plain aquifer system; therefore, smectite is the likely product of plagioclase weathering reactions.

Additional evidence for incongruent dissolution of plagioclase is the association of basalt and montmorillonite. Wood and Low (1988, p. D5, D10) observed both an iron-rich green smectite and an iron-poor white montmorillonite as alteration products of basalt from the Snake River Plain aquifer system. Rightmire and Lewis (1987a, p. 59-70) also observed the presence of secondary smectite and mixed-layer clays that were isolated in basalt vesicles. The ubiquitous occurrence of plagioclase and the limited occurrence of associated weathering products in the Snake River Plain aquifer suggest that this mineral is actively weathering in this system, but the amount of weathering may be small when compared with olivine.

Another indication that plagioclase weathering may be a limited process in the

Snake River Plain aquifer is the aqueous isotopic ratio of $^{87}\text{Sr}/^{86}\text{Sr}$. Leeman (1982, fig. 4) noted $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios of 0.706 to 0.707 for olivine basalts of the Snake River Plain aquifer system. Plagioclase commonly contains trace amounts of strontium substituting for calcium in the mineral lattice (Verhoo-gen and others, 1970, p. 80) and is a potential source of dissolved strontium in Snake River Plain aquifer water. Leeman (1982, table 2) reported strontium concentrations in olivine basalts of the Snake River Plain aquifer system of about 200 to 300 ppm, and Knobel and others (1995, p. 49) reported a mean strontium concentration of 296 ppm for 84 basalt samples from the INEL. If plagioclase were the primary source mineral for dissolved strontium in Snake River Plain aquifer water, then the isotopic ratio of $^{87}\text{Sr}/^{86}\text{Sr}$ would be similar to that of the basalt. Observed $^{87}\text{Sr}/^{86}\text{Sr}$ values in the Snake River Plain aquifer water samples were from 0.709 to 0.711 (R.J. Hunt, U.S. Geological Survey written commun., 1993). The larger isotopic ratios in the dissolved strontium fraction suggest that plagioclase feldspar dissolution may be a relatively minor process in the Snake River Plain aquifer.

The molar composition of plagioclase feldspar was determined for one basalt sample by Wood and Low (1988, p. D8) to be equal in calcium and sodium. This composition is in the range of the plagioclase mineral labradorite. Labradorite also was identified in a RWMC sediment sample by Rightmire (1984, p. 21) and contained a molar calcium to sodium ratio of 0.6:0.4. Kuntz and Dalrymple (1979, p. 18) also found plagioclase in the chemical composition range of labradorite: 0.65 calcium and 0.35 sodium to 0.5 calcium and 0.5 sodium. Because of the variability in labradorite compositions, an idealized chemical equation ($\text{Ca}_{0.5}\text{Na}_{0.5}\text{Al}_{1.5}\text{Si}_{2.5}\text{O}_8$) was used in this report.

Labradorite dissolves in water when CO_2 is present to form calcium montmorillonite and

dissolved silica, sodium, calcium, and bicarbonate. A reaction model for the incongruent dissolution of labradorite (equation 5) is provided in table 5.

Precipitation of Calcite

Calcite is widely distributed in the Snake River Plain aquifer system. Surficial sediments contain from 5 to 15 percent calcite, and calcite is the predominant mineral in interbed samples from the TAN region (Bartholomay, 1990, table 2, p. 9). Calcite also fills vesicles and fractures in basalt (Rightmire and Lewis, 1987a, p. 59-70; Wood and Low, 1988, fig. 10).

The saturation indices discussed previously indicate that Snake River Plain aquifer water samples are supersaturated with respect to calcite (fig. 9). As a result, precipitation reactions are the only thermodynamic possibility. Generally, stoichiometric calcite precipitates unless $[Mg^{+2}]:[Ca^{+2}]$ is greater than one (brackets indicate ion activities). If the ratio is greater than 1 calcite can be altered to dolomite; however, conversion at low temperatures is rare unless the ratio is considerably larger than one (Drever, 1988, p. 66). If the ratio is near that of sea water (about 5.2), calcite can contain 11 to 19 mole percent magnesium carbonate and the resultant mineral is high-magnesium calcite (Drever, 1988, p. 66). Ratios of $[Mg^{+2}]:[Ca^{+2}]$ were calculated for the data tabulated by Knobel and others (1992, table 5). The mean and median values for 24 Snake River Plain aquifer samples were 0.63 and 0.60, respectively. These data indicate that stoichiometric calcite should be the carbonate mineral precipitated from solution in this aquifer.

When water that is saturated with respect to calcite comes into contact with sufficient numbers of solid-phase nucleation sites, dissolved calcium and bicarbonate combine to form calcite, dissolved CO_2 , and water. Dissolved CO_2 was observed by Busenberg and

others (1993, table 1) in nine ground-water samples from wells on or near the INEL. Concentrations of dissolved CO_2 were from 1.5 to 5.6 mg/L and were distributed about mean and median concentrations of 3.9 and 4.3 mg/L, respectively. All Snake River Plain aquifer wells included in the report by Knobel and others (1992) contained water with calculated CO_2 partial pressures (Plummer and others, 1978) larger than atmospheric (table 6). The CO_2 partial pressures were from 1.35 times standard air to 14.5 times standard air. These data are consistent with CO_2 generation during the precipitation of calcite.

Additional evidence for the precipitation of calcite in the Snake River Plain aquifer system is the direct observation of secondary calcite by thin-section analysis (Rightmire and Lewis, 1987a, p. 59-70) and visual examination (Wood and Low, 1988, fig. 10). A reaction model for the precipitation of pure calcite (equation 6) is provided in table 5.

Dissolution-Precipitation Reactions Involving Silica

Silica occurs in a variety of structural forms such as crystalline quartz, cryptocrystalline chalcedony, and amorphous silica glass. Quartz makes up 30 to 40 percent of surficial sediment samples and 20 to 40 percent of sedimentary interbed samples. It is the most abundant mineral in these unconsolidated materials. In basalts, some detrital quartz is observed as fracture- and vesicle-fill material, however, quartz generally does not occur as a primary mineral. Amorphous silica glass that is free of inclusions is rare in Snake River Plain basalt flows and is intersertal in most samples (Lanphere and others, 1993, p. 19). Amorphous silica glass should not be confused with tachylyte or the fine-grained groundmass that is common in the Snake River Plain basalt flows. The fine-grained groundmass essentially is made up of smaller crystals of the same

minerals as the rest of the basalt (Lanphere and others, 1993, p. 19).

The theoretical controls on the concentration of dissolved silica (SiO_2) in ground water are the solubilities of quartz (lower limit) and amorphous silica (upper limit). The equilibrium constants at 25°C are 0.0001 for quartz and 0.002 for amorphous silica (Drever, 1988, p. 99). These values correspond to concentrations of 6 and 120 mg/L, respectively. The actual concentrations of silica in 23 water samples from Snake River Plain aquifer wells were within these limits. Concentrations were from 19 to 35 mg/L (Knobel and others, 1992, table 5). The limited occurrence of amorphous silica in the Snake River Plain basalt flows and the fact that SiO_2 concentrations are less than 120 mg/L suggest that some mechanism other than dissolution of amorphous silica may be controlling the upper limit of silica concentrations in ground water. Although it is unknown what the solubility control is, a likely possibility is that actual concentrations may be related to the rates and mechanisms of the irreversible weathering reactions of the complex aluminosilicate minerals (Garrels, 1976, p. 75).

Saturation indices (fig. 11) calculated for water samples tabulated by Knobel and others (1992) indicate that water from the Snake River Plain aquifer is supersaturated with respect to the silica minerals quartz and chalcedony; hence, precipitation of these minerals is the only thermodynamic possibility. Conversely, water samples are slightly undersaturated with respect to amorphous silica (fig. 11), and dissolution should take place.

Silica reactions in the Snake River Plain aquifer system probably are complex and varied. For example, Wood and Low (1988, p. D5-D8 and table 3) observed cryptocrystalline silica as vesicle linings in deep basalts (68-325 m) and as amorphous silica in sedimentary material. Rightmire and Lewis (1987a, p. 59-70) observed amorphous silica as grain coatings, fracture fill, and vesicle fill

in the unsaturated zone at the RWMC. Silica in the crystalline form was observed in the outcrop (Wood and Low, 1988, p. D8) and in the unsaturated zone (Rightmire and Lewis, 1987a, p. 60). These observations coupled with the thermodynamic data (fig. 11) imply that silica precipitates directly from solution in the saturated zone (cryptocrystalline and/or crystalline) and as a result of solution concentration resulting from evaporation in the unsaturated zone (amorphous).

Secondary amorphous silica can also be unstable as indicated by calcite replacement of silica in the unsaturated zone (Rightmire and Lewis, 1987a, p. 60). Alteration of glass in basalt to a mass of opaque minerals and pyroxene (Lanphere and others, 1993, p. 20) suggested a quasi-incongruent dissolution of amorphous silica in the Snake River Plain aquifer system.

Fortunately, silica reactions are stoichiometrically simple, and all of the reactions can be represented by a single reaction model. Equation 7 (table 5) represents the dissolution or precipitation of either crystalline, cryptocrystalline, or amorphous silica. The saturation indices calculated by WATEQF (Plummer and others, 1978) and shown in figure 11 are for the dissolution reactions.

Oxidation of Ferrous Iron

Ferrous iron is contained in olivine (table 5) and pyroxene (Wood and Low, 1988, table 10). It is also the form of iron present in pyrite. If these minerals dissolve, ferrous iron is released to solution. In the presence of oxygen, ferrous iron is unstable and oxidizes to ferric iron. Ferric iron immediately reacts with oxygen and water to form hydrogen ions and solid ferric hydroxide or one of its precursory phases such as ferric oxyhydroxide. Sufficient dissolved oxygen for oxidation reactions to proceed was noted in the Snake River Plain aquifer samples listed in the report by Knobel and others (1992, table 3) and in INEL soil-gas

samples discussed in the report by Rightmire and Lewis (1987b, table 12).

The oxidation of ferrous to ferric iron is an inherent part of the reaction model for the dissolution of olivine (equation 2) given in table 5. The reaction model for the dissolution of diopside (equation 3) does not account for any ferrous iron that may be contained in that mineral. As a result, a separate reaction model (equation 8) is necessary to account for any ferrous iron that may be present in diopside or other minerals such as pyrite (table 5).

The iron weathering rinds associated with basalt minerals, especially olivine (Lanphere and others, 1993, p. 20), are additional evidence that ferric iron forms from weathering of olivine and precipitates from solution. Pyrite also has been observed in Snake River Plain aquifer system (Crosthwaite, 1973; Doherty, 1979) and may contribute ferrous iron to the system. A reaction model for the dissolution of pyrite (equation 9) is provided in table 5.

COMPARISON OF RESULTS WITH RESULTS OF PREVIOUS INVESTIGATIONS

Results of several studies of Snake River Plain aquifer system geochemistry previously have been reported in the scientific literature. These studies included concepts that were partly duplicated in this study. The areal scope was either larger (Robertson and others, 1974; Wood and Low, 1988) or smaller (Rightmire and Lewis, 1987b; Hull, 1989; Rawson and Hubbell, 1989; Rawson and others, 1991) than the scope of this study, or it was the same as this study (Olmsted, 1962). This section provides—in chronological order—a comparison of the results of previous work with the results with this study.

Olmsted, 1962.—His study describes the chemical and physical properties of ground water beneath the National Reactor Testing

Station, which is now known as the INEL. Olmsted (1962, table 1) tabulated the results of 148 chemical analyses of ground water from the INEL and vicinity. Olmsted (1962, tables 2-6) listed calculations of percentage reacting values for major anions and cations and assigned a letter designating the type of water for each sample analyzed (Olmsted, 1962, tables 1-6).

Olmsted (1962, p. 18-19, and fig. 2) classified the water beneath the INEL into four chemical types, designated A, B, C, and D. Type A and B waters contain a cation composition that is predominantly calcium and magnesium and an anion composition that is predominantly bicarbonate and carbonate. Type A and B waters are equivalent to the calcium-magnesium bicarbonate water identified in the present study (fig. 6) for water samples isolated from waste-disposal sites. Type C and D waters have no limits set on the cation composition; however, the required anion compositions are greater than 30 percent chloride for type C water and greater than 30 percent sulfate for type D water. In the present study, several samples from TAN (fig. 7) and from the RWMC (fig. 8) were enriched with chloride and generally correlate to Olmsted's type C water.

In addition, Olmsted (1962, p. 69) studied the areal variations in chemical character of ground water and concluded that the variations mimic the ground-water flow system. This conclusion is in agreement with the present study. The variations in chemical character were attributed to the processes of dilution, sorption, and chemical precipitation. Although dilution (mixing of waters) was not addressed in the present study, sorption and mineral precipitation are consistent with the reaction models hypothesized during this study.

Olmsted (1962, p. 24-31) noted that water from wells not located near waste-discharge facilities had minimal water-chemistry changes with both depth and time. However,

water from some wells near waste-disposal sites did show variations with depth and time. No attempt to describe vertical and temporal changes in water chemistry was made in the present study.

Robertson and others, 1974.—The emphasis of their study is the influence of waste disposal on geochemistry of water at the National Reactor Testing Station. Their study includes an analysis of saturation indices of water samples with respect to Snake River Plain aquifer minerals and identifies reactions taking place in the recharge areas contributing water to the Snake River Plain aquifer.

Ground-water samples from and adjacent to the INEL were found to be saturated with respect to calcite and dolomite (Robertson and others, 1974, p. 77-79). These results are consistent with the results of the present study (fig. 9). Robertson and others (1974, p. 79-81) observed that water samples from the Snake River Plain aquifer were undersaturated with respect to magnesian serpentine (a surrogate mineral used to approximate the thermodynamic state of olivine). For comparison purposes, the present study indicates that forsterite (an olivine mineral) is unstable with respect to Snake River Plain aquifer water samples (fig. 11). Although Robertson and others (1974, p. 79) used an incongruent hydrolysis reaction to evaluate olivine stability, a more likely reaction is the dissolution of olivine in the presence of CO_2 and O_2 (equation 2, table 5). Regardless of the approach taken, both methods of analysis lead to the conclusion that olivine is unstable with respect to water samples taken from the Snake River Plain aquifer on or adjacent to the INEL.

Robertson and others (1974, p. 42-58) discussed hypothetical reactions occurring in the recharge areas surrounding the INEL. Many of the reactions proposed were hydrolysis reactions involving silicate, aluminosilicate, and carbonate minerals. Other reactions included dissolution of sulfate and fluoride

minerals and oxidation of iron sulfides and ferrous iron. These reactions are not directly comparable to the results of the present study because the lithology and mineralogy of the source rocks in the recharge areas differ from the Snake River Plain aquifer geologic materials. However, some of the reactions are similar to reactions identified in the present study for the Snake River Plain aquifer system at the INEL. Examples are the dissolution of anhydrite (eq. 4, table 5) and oxidation of iron (eqs. 8-9, table 5). Hydrolysis reactions (Robertson and others, 1974) differ somewhat from silicate and carbonate mineral reactions included in table 5 because the atmospheric components CO_2 and O_2 were included in reactions considered in the present study.

Rightmire and Lewis, 1987b.—The objective of the study by Rightmire and Lewis (1987b) was to develop a conceptual model of the geochemical environment of the shallow unsaturated zone at the RWMC (Rightmire and Lewis, 1987b, p. 80). As part of their study, saturation indices were calculated for three Snake River Plain perched-water samples relative to several minerals (Rightmire and Lewis, 1987b, table 16). The results of that study are not directly comparable to the results of the present study because perched-water chemistry is not considered in the present study. Median values of saturation indices calculated by Rightmire and Lewis (1987b, table 16) indicated that the water samples were supersaturated with respect to calcite, chalcedony, and quartz. These data are consistent with the results of the present study. Conversely, the perched-water samples were oversaturated with respect to strontionite. This conclusion conflicts with the finding of the present study that Snake River Plain aquifer samples are undersaturated with respect to strontionite. Because strontionite has not been observed in the geologic materials underlying the Snake River Plain, undersaturation of Snake River Plain aquifer water is more likely than oversaturation; however, oversaturation in

perched-water bodies may result from waste-disposal practices.

Wood and Low, 1988.—In a comprehensive study of the solute geochemistry of the Snake River Plain Regional aquifer system, Wood and Low (1988) used several geochemical techniques and concepts that also are used in the present study. Specifically, they used an analysis of ion distribution and thermodynamics as part of their formulation of a proposed set of chemical reactions that described the solute geochemistry of the eastern Snake River Plain aquifer system.

Wood and Low (1988, table 20A) tabulated water-chemistry data for the Snake River Plain aquifer system. Data for 424 analyses of eastern Snake River Plain aquifer samples were extracted for use in the present study and mean concentrations were computed (table 4). Those data were compared with data collected for the present study and were discussed previously in this report.

Wood and Low (1988, table 13) also computed saturation indices for six ground-water samples with respect to eight minerals. The water samples were collected from the eastern Snake River Plain aquifer near the discharge area between Twin Falls and King Hill. The water samples were undersaturated with respect to anhydrite, diopside, and forsterite. In addition, the water samples were near equilibrium with respect to calcite and silica glass. These findings generally are in agreement with the present study. The saturation indices calculated by Wood and Low (1988, table 13) for the water samples with respect to albite, anorthite, and calcium smectite indicated undersaturation for albite and anorthite and saturation for calcium smectite. Although saturation indices for reactions involving these minerals were not calculated in the present study because of uncertainties in the thermodynamic constants associated with the minerals, analysis of mineral stability diagrams for the same minerals (figs. 15-20) indicate direct

comparability with the findings of Wood and Low (1988).

The set of chemical reactions controlling solute geochemistry of the eastern Snake River Plain aquifer system that was proposed by Wood and Low (1988, table 10) is nearly identical to the preliminary reaction models proposed in the present study (table 5) for the eastern Snake River Plain aquifer system in the vicinity of the INEL. The small discrepancies result from minor differences in the mineral stoichiometries used for constructing the reaction models.

Hull, 1989.—A conceptual model was developed to describe how infiltrating warm-waste pond water at the TRA (fig. 1), affects the environment. Hull (1989, fig. 18) compared hydrochemical facies of water from the Snake River Plain aquifer with hydrochemical facies of water from a perched-water body beneath the pond. The hydrochemical-facies results for Snake River Plain aquifer samples discussed by Hull (1989) are in agreement with the hydrochemical facies for aquifer-water samples from locations away from the RWMC and TAN that are discussed in the present study (fig. 6).

Rawson and others, (1989, 1991).—The results of a study by Rawson and her colleagues is reported by Rawson and Hubbell (1989) and Rawson and others (1991). The purpose of their study was to determine the geochemical controls on the composition of soil pore waters in the unsaturated zone beneath a mixed-waste disposal site at the RWMC and to determine the extent of radionuclide migration from the disposal site. Major-ion concentrations were determined for pore-water samples (Rawson and others, 1991, table 2); hydrochemical facies were plotted on Piper diagrams (Rawson and Hubbell, 1989, fig. 6); and saturation indices of the pore waters were calculated with respect to selected minerals (Rawson and Hubbell, 1989, p. 245). The results of these studies differ from the

results of the present study because pore-water chemistry and aquifer-water chemistry are not comparable. The incomparability of similar types of chemical data (pore-water samples from the saturated zone and ground-water samples) was established by studies conducted in the northern Atlantic Coastal Plain aquifer system (Trapp and others, 1984, table 17; Pucci and others, 1992, p. 426).

EFFECTS OF WASTE DISPOSAL AT SELECTED SITES

Natural geochemical reactions in the Snake River Plain aquifer system locally may be modified by the disposal of wastes to the sub-surface at INEL facilities. Waste-disposal practices at the INEL have included wastewater discharge to deep injection wells and infiltration ponds, and the burial of solid and liquid wastes. These different methods of waste disposal have affected the concentrations, vertical distribution, and horizontal extent of contaminants in ground water. Deep injection wells at TAN, ICPP, and TRA provided point-source disposal that introduced wastewater contaminants directly to the Snake River Plain aquifer over a broad vertical interval of aquifer material. The use of these deep injection wells has been discontinued. In deep injection, less conservative contaminants were injected directly to the aquifer at waste-stream concentrations, and sorption and decay processes were initiated within the aquifer rather than in the unsaturated zone. Infiltration ponds at TAN, ICPP, and TRA have been used for disposal of wastewater containing radionuclides, inorganic and organic chemical constituents, and sanitary wastes. Water percolating from infiltration ponds must pass through a thick sequence of unsaturated or partially saturated basalt flows and interbeds before reaching the aquifer. Movement through the unsaturated zone permits sorption and radioactive decay prior to contact with the aquifer.

Recharge takes place at the top of the Snake River Plain aquifer and is diffused over a comparatively large area. In addition, small hydraulic gradients and small ratios of vertical-to-horizontal hydraulic conductivity restrict the vertical movement of wastewater contaminants to the upper part of the aquifer. Solid and liquid wastes have been buried in trenches and pits at the RWMC since the early 1950's. The mobilization of radionuclides, inorganic chemicals, and purgeable organic chemicals in these buried wastes largely is dependent on occasional flooding of the RWMC during periods of excessive local runoff.

Test Area North

The TAN disposal well was used during 1953-72 to dispose of wastewater containing low-level radionuclides, inorganic and organic chemical constituents, and sanitary wastewater. Water-quality monitoring at the 310-ft-deep TAN disposal well was begun after waste injection was discontinued. A 35-acre infiltration pond at TAN was constructed in 1972 to replace the TAN disposal well. The pond is used to dispose of wastewater containing radionuclides, inorganic and organic chemical constituents, and sanitary wastes.

During 1959-91, about 61 Ci of radioactivity was discharged in wastewater to the disposal well and infiltration pond. Of this amount, approximately 20 Ci was discharged to the disposal well in 1968 and 1969 in response to problems with an evaporator used to reduce the volume of liquid waste (Energy Research and Development Administration, 1977, p. II-110, II-111). Use of the evaporator was phased out in 1969.

During 1989-91, the average volume of radioactive wastewater discharged to the infiltration pond at TAN was about 18.4 million gal/year. The average rate of disposal of radioactivity in this wastewater was 0.061 Ci/year.

An average of approximately 27.9 million gal/year of chemical wastes was discharged to the infiltration pond during 1989–91. The predominant constituents were chloride and sodium. Average annual amounts of 11,500 lb of chloride and 7,500 lb of sodium were discharged. The average annual amount of all other chemical constituents in the effluent was about 6,000 lb (Bartholomay and others, 1995, p. 17).

Radionuclides, including cobalt-60, cesium-137, plutonium isotopes, and americium-241 were detected in samples collected from the TAN disposal well in 1987–88 (Knobel and Mann, 1988, table 3). Other than at the TAN injection well, no radionuclide or inorganic-chemical plume has been detected near TAN; however, purgeable organic compounds, including trichloroethylene, (Mann and Knobel, 1987, table 2; Mann, 1990, table 2) have been detected in water from wells near TAN.

Idaho Chemical Processing Plant and Test Reactor Area

Since 1952, the chemistry of ground water at the INEL has been affected most measurably over a large area by wastewater-disposal practices at the ICPP and TRA. The deep injection well at the ICPP was used until 1984 to dispose of most wastewater containing radionuclides and inorganic chemical constituents. Since 1984, two infiltration ponds south of the ICPP have been used for disposal of wastewater. The TRA injection well was used during 1964–82 to dispose of cooling-tower wastewater containing inorganic chemical constituents derived from anticorrosive additives. TRA wastewater containing radionuclides, chemical constituents, and sanitary wastes has been discharged since 1952 to a series of infiltration ponds.

Deep injection wells.—Tritium was the predominant radionuclide in wastewater

disposed to the ICPP deep injection well (Orr and Cecil, 1991, p. 20). Although some of the tritium was disposed to infiltration ponds at the TRA (1952–91) and at the ICPP (1984–91), approximately two-thirds of the total tritium were disposed to the ICPP deep injection well. Approximately 20,000 Ci of tritium were disposed to the well during 1952–84. Since disposal began, tritium concentrations in water from wells downgradient from the ICPP have greatly exceeded background tritium concentrations. The measurable plume consists of two plumes that merged. One plume was derived primarily from the ICPP deep injection well and the other from wastewater infiltrating from ponds at the TRA. In 1991, the area of the measurable tritium plume with concentrations exceeding 0.5 pCi/mL was about 40 mi² (Bartholomay and others, 1995, p. 22). Although the tritium plume is laterally extensive downgradient from the ICPP, the effects on geochemical reactions within the aquifer probably are minimal because tritium, as an isotope of hydrogen, largely is tied up in water molecules and behaves chemically like stable hydrogen.

Wastewater disposed to the ICPP deep injection well also contained small amounts of strontium-90. In 1991, the area of a plume containing strontium-90 at concentrations exceeding 5 pCi/L was less than 1 mi². The area of the strontium-90 plume has decreased in response to decreases in disposal rates, sorption in the aquifer, sorption in the unsaturated zone following the change in method of disposal from deep injection well to infiltration pond, and radioactive decay. Strontium-90 is expected to be strongly sorbed to the aquifer-system matrix and locally may affect geochemical reactions in the aquifer. Small quantities of other radionuclides, including plutonium isotopes and cesium-137, also were disposed to the ICPP deep injection well. Until the middle and late 1980's, concentrations of these radionuclides exceeded the reporting level in water from two wells just downgradient from the ICPP deep injection well.

Plutonium isotopes were not detected in other wells downgradient from the ICPP deep injection well.

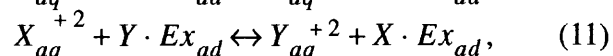
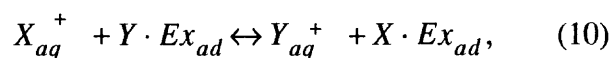
Inorganic chemical constituents were disposed to the ICPP deep injection well during 1952-84 and, to a lesser degree, to the TRA injection well during 1964-82. Sodium, chloride, sulfate, nitrate, and fluoride were the predominant inorganic constituents in wastewater disposed to the ICPP deep injection well; sulfate and chromium were the predominant constituents in wastewater disposed to the TRA injection well.

Measurable plumes of sodium, chloride, and nitrate formed downgradient from the ICPP deep injection well (Orr and Cecil, 1991, p. 40-44). In 1991, sodium concentrations in water from wells downgradient from the ICPP exceeded 70 mg/L, as much as seven times larger than sodium concentrations in areas unaffected by wastewater disposal. In a similar fashion, chloride concentrations exceeded 160 mg/L, more than eight times larger than chloride concentrations in unaffected areas. Although large amounts of sulfate have been discharged to the injection wells at the ICPP and TRA, detectable sulfate concentrations in ground water have not increased in either area. In a general sense, these constituents are nonreactive in the Snake River Plain aquifer; however, the disposal of large volumes of these constituents locally may disrupt the geochemical equilibrium of the natural flow system.

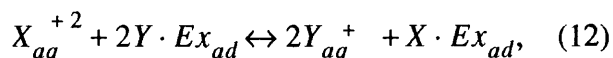
Infiltration ponds.—At the TRA, a sequence of ponds have been used for disposal of wastewater containing radionuclides since 1952. In 1962, the chemical-waste pond was constructed and nonradioactive wastewater containing industrial chemicals was diverted to it. Disposal to the TRA injection well was discontinued when the cooling-tower wastewater was diverted to cold-waste infiltration ponds constructed in 1982. An unlined sanitary-waste pond has received all sanitary waste-

water since the early 1950's. The infiltration ponds at the ICPP were constructed in 1984 to replace the ICPP injection well.

Perched ground-water bodies have formed in the unsaturated basalt and sedimentary interbeds beneath the infiltration ponds at the TRA and ICPP. Water in these perched bodies contains large concentrations of radionuclides and inorganic chemicals. Radionuclide and chemical constituents in wastewater discharged to the ponds infiltrate the pond bottoms. More conservative constituents move through a series of perched ground-water bodies and unsaturated basalt and sediment before reaching the aquifer and contributing to contaminant plumes. Concentrations of less conservative constituents may not be detectable in water from the aquifer because of retardation occurring in the unsaturated zone. One effective process for such retardation is cation exchange. The process can be represented by the equations:



and



where

X_{aq}^{+} and Y_{aq}^{+} = dissolved monovalent cations

X_{aq}^{+2} and Y_{aq}^{+2} = dissolved divalent cations, and

$X \cdot Ex_{ad}$ and $Y \cdot Ex_{ad}$ = cations adsorbed onto an exchange substrate.

This exchange occurs stoichiometrically, so that electrical neutrality is maintained both on the exchanging surface and in the water. The process was described in detail by Chapelle and Knobel (1983). The net effect of cation exchange is that the multivalent, heavier cations found in waste streams tend to be removed from solution by adsorption and replaced by lower-charged cations that are less

stable on solid-phase exchange sites. The capacity of solid-phase material to exchange cations with a solution is easy to measure and is reported by laboratories as cation exchange capacity (CEC) in milliequivalents per 100 g of dry weight. Bartholomay and others (1989, table 14) summarized CEC data collected at the INEL by the USGS prior to 1989. The CEC's they tabulated are sufficiently large to provide a significant retardation effect on liquid wastes disposed to infiltration ponds at the INEL. Although amounts of strontium-90 disposed to the warm-waste ponds at TRA were comparable to the amounts disposed to the ICPP injection well, no strontium-90 plume has been identified from TRA disposal. The lack of a strontium-90 plume at TRA probably results from cation exchange in the unsaturated zone.

Radioactive Waste Management Complex

Solid and liquid wastes have been buried in trenches and pits in surficial sediments at the Subsurface Disposal Area (SDA) at the RWMC since the early 1950's. About 550 Ci of plutonium-238, 21,000 Ci of plutonium-239, 4,900 Ci of plutonium-240, 165,000 Ci of plutonium-241, and 51,000 Ci of americium-241 were buried in the SDA during 1954–70 (Barraclough and others, 1976, p. 11). Organic compounds also have been buried. An estimated 88,400 gal of organic waste disposed prior to 1970 contained about 24,400 gal of carbon tetrachloride, 39,000 gal of lubricating oil, and about 25,000 gal of other organic compounds, including trichloroethane, trichloroethylene, perchloroethylene, toluene, and benzene (Mann and Knobel, 1987, p. 1).

Mobilization of waste constituents, including transuranic wastes, other radionuclides, chemical constituents, and purgeable organic compounds, is dependent in part on availability of recharge water from local flood

events. Flooding of the SDA occurred in 1962 and 1969. Water infiltrating the surficial sediments during and after flooding may have provided a mechanism for transport of contaminants downward to the Snake River Plain aquifer. Trench construction also may affect mobilization of waste constituents. Prior to 1970, little or no sediment was retained between the trench bottoms and underlying basalt. Since 1970, a layer of sediment has been retained to inhibit downward migration of constituents. Trench caps also may affect the mobilization of these constituents.

Concentrations of americium-241 have been detected in wells near the RWMC. Americium-241 is a decay product of plutonium-241, which has a half-life of 14.4 years. Plutonium isotopes and americium-241 have been buried at the RWMC.

Sodium, chloride, and nitrate concentrations in ground water near the RWMC historically have exceeded background concentrations. In 1991, sodium, chloride, and nitrate concentrations were as large as 45, 90, and 8.0 mg/L, respectively, in water from wells adjacent to the RWMC (Bartholomay and others, 1995, p. 31-33).

Purgeable organic compounds have been detected in wells at the RWMC (Mann and Knobel, 1987, table 2; Mann, 1990, table 2; Liszewski and Mann, 1992, table 2). In 1990, for example, concentrations of carbon tetrachloride and trichloroethylene in well 88 were as large as 3.9 and 1.3 µg/L, respectively (Liszewski and Mann, 1992, table 2).

SUMMARY AND CONCLUSIONS

This report delineates the natural geochemical reactions that partially control the chemistry of ground water in the Snake River Plain aquifer in southeastern Idaho at the INEL. The report was prepared by the USGS in cooperation with the DOE's Idaho Operations Office.

A mineralogical and mineral-stability analysis of the solid-phase materials of the Snake River Plain aquifer system revealed that labradorite, pyroxene, and olivine are the major unstable minerals in this aquifer system, whereas smectite, calcite, and ferric oxyhydroxide are the major stable minerals. Silica phases (quartz, cryptocrystalline quartz, and amorphous silica) can be either stable or unstable. In basalt samples, the relative amounts of major minerals in decreasing order of abundance were plagioclase feldspar, pyroxene, and olivine. Minerals directly observed as secondary weathering products in association with basalt or with minerals in basalt included smectite, ferric oxyhydroxide, calcite, and silica. Calcite is an important component of surficial materials and sedimentary interbeds, and anhydrite may be present.

Soluble minerals in the aquifer system interact with ground water to add dissolved constituents to solution. Concentrations of dissolved constituents tend to increase in the generalized direction of ground-water flow from relatively more dilute solutions at the INEL to less dilute solutions where the ground water discharges at springs along the Snake River. Mean calcium, magnesium, sodium, potassium, bicarbonate, chloride, sulfate, and silica concentrations changed from 40 to 50 mg/L, 15 to 20 mg/L, 12 to 33 mg/L, 2.6 to 4.6 mg/L, 177 to 216 mg/L, 14 to 38 mg/L, 21 to 48 mg/L, and 29 to 34 mg/L, respectively. These changes reflect, in part, the increased amount of time that ground water has been in contact with soluble minerals in the aquifer system.

The hydrochemical facies of water samples are useful tools for describing the chemical character of ground water. Water samples from the Snake River Plain aquifer south of NRF generally were calcium-magnesium bicarbonate in character. Water samples from TAN had larger relative concentrations of sodium and chloride and smaller relative concentra-

tions of calcium and bicarbonate than other water samples considered in this report. The differences in water chemistry at TAN are attributed to waste-disposal practices.

The link between the mineral assemblage of the Snake River Plain aquifer system (solid phases) and the aqueous solution (dissolved gases, water, and hydrated ions) is the energy level or thermodynamic condition of the chemical system. Saturation indices calculated for this study from 22 representative water samples indicate that anhydrite, pyroxene (diopside), olivine, and possibly amorphous silica are unstable and should dissolve. Water is oversaturated with respect to calcite and it should precipitate. Because ferric iron is unstable in the presence of oxygen, it precipitates as a precursory phase to ferric hydroxide. Mineral-stability diagrams for the complex aluminosilicate minerals indicate that labradorite can weather to form calcium montmorillonite in this aquifer system.

Plausible chemical reactions in this report are chemical reactions that are likely to take place naturally in the Snake River Plain aquifer system because (1) the requisite source minerals, liquids, and gases are present in the aquifer system; (2) thermodynamic conditions allow the reaction to proceed; and (3) the dissolved, gaseous, or solid-product phases either are present in or can be accounted for by processes that remove them from the aquifer system. These reactions include congruent dissolution of olivine, diopside, and anhydrite; incongruent dissolution of labradorite with residual calcium montmorillonite; precipitation of calcite; oxidation of ferrous iron and precipitation of ferric oxyhydroxide; and either precipitation or dissolution of silica.

Waste-disposal practices modify the natural chemistry in the Snake River Plain aquifer system, but the rates and amounts of modification are dependent on the type of disposal practice. For example, when wastes are injected directly into the saturated part of the

system, they bypass many of the natural geochemical barriers that would immobilize some of the waste constituents; hence, they arrive in the aquifer at waste-stream concentrations. Conversely, when wastes are discharged to infiltration ponds, many constituents are retarded during downward infiltration of waste fluid. Cation exchange is an effective process for such retardation.

The net effect of cation exchange is that the multivalent, heavier cations in waste streams tend to be removed from solution by adsorption and replaced by lower-charged cations that are less stable on solid-phase exchange sites. The cation-exchange capacities of aquifer-system materials are sufficiently large to provide a significant retardation effect on liquid wastes discharged to infiltration ponds at the INEL.

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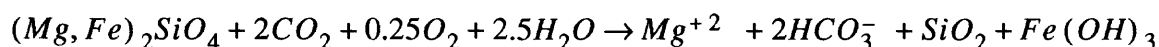
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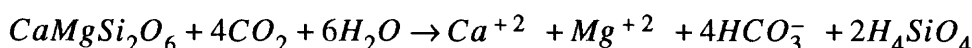
Table 5—Preliminary reaction models used for interpreting the natural geochemistry of the Snake River Plain aquifer system, Idaho

CONGRUENT DISSOLUTION REACTIONS

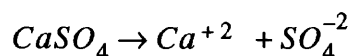
(2) Dissolution of olivine



(3) Dissolution of diopside

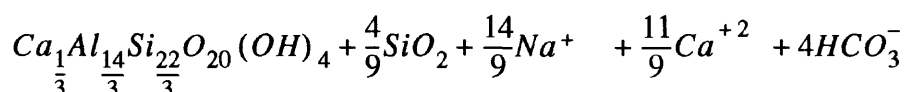
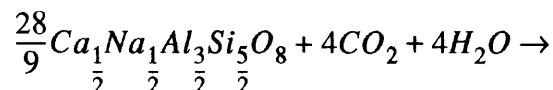


(4) Dissolution of anhydrite



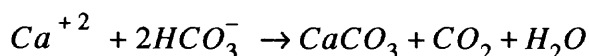
INCONGRUENT DISSOLUTION OF FELDSPAR

(5) Weathering of labradorite to calcium montmorillonite (calcium smectite)

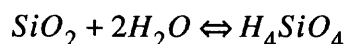


PRECIPITATION-DISSOLUTION REACTIONS

(6) Precipitation of calcite

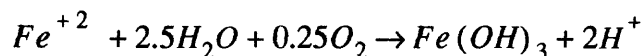


(7) Silica dissolution and/or precipitation



OXIDATION-REDUCTION REACTIONS

(8) Oxidation of ferrous iron



(9) Oxidation of pyrite

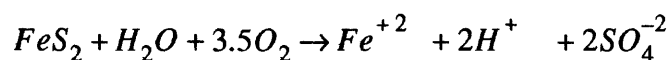


Table 6—Calculated equilibrium partial pressures of carbon dioxide dissolved in water from selected Snake River Plain aquifer wells, from Webb Spring, and in standard air

[Partial pressures were calculated using the computer program WATEQF (Plummer and others, 1978). Sample identifier, data, and locations are given by Knobel and others (1992)]

Sample identifier	Carbon dioxide (atmospheres $\times 10^{-4}$)
[Atmosphere]	
Standard air	3.1
[Radioactive Waste Management Complex wells]	
USGS 87	9.6
88	6.0
89	4.2
90	9.0
117	5.2
119	4.7
120	12
RWMC PROD.	11
[Test Area North wells]	
TAN DISP.	22
TDD-1	12
TDD-2	26
TDD-3	22
IET DISP.	26
ANP-8	7.7
USGS 24	18
[Selected wells and spring]	
Arbor Test	9.3
NRF-2	45
USGS 11	12
14	9.2
98	22
108	11
Webb Spring	0.47