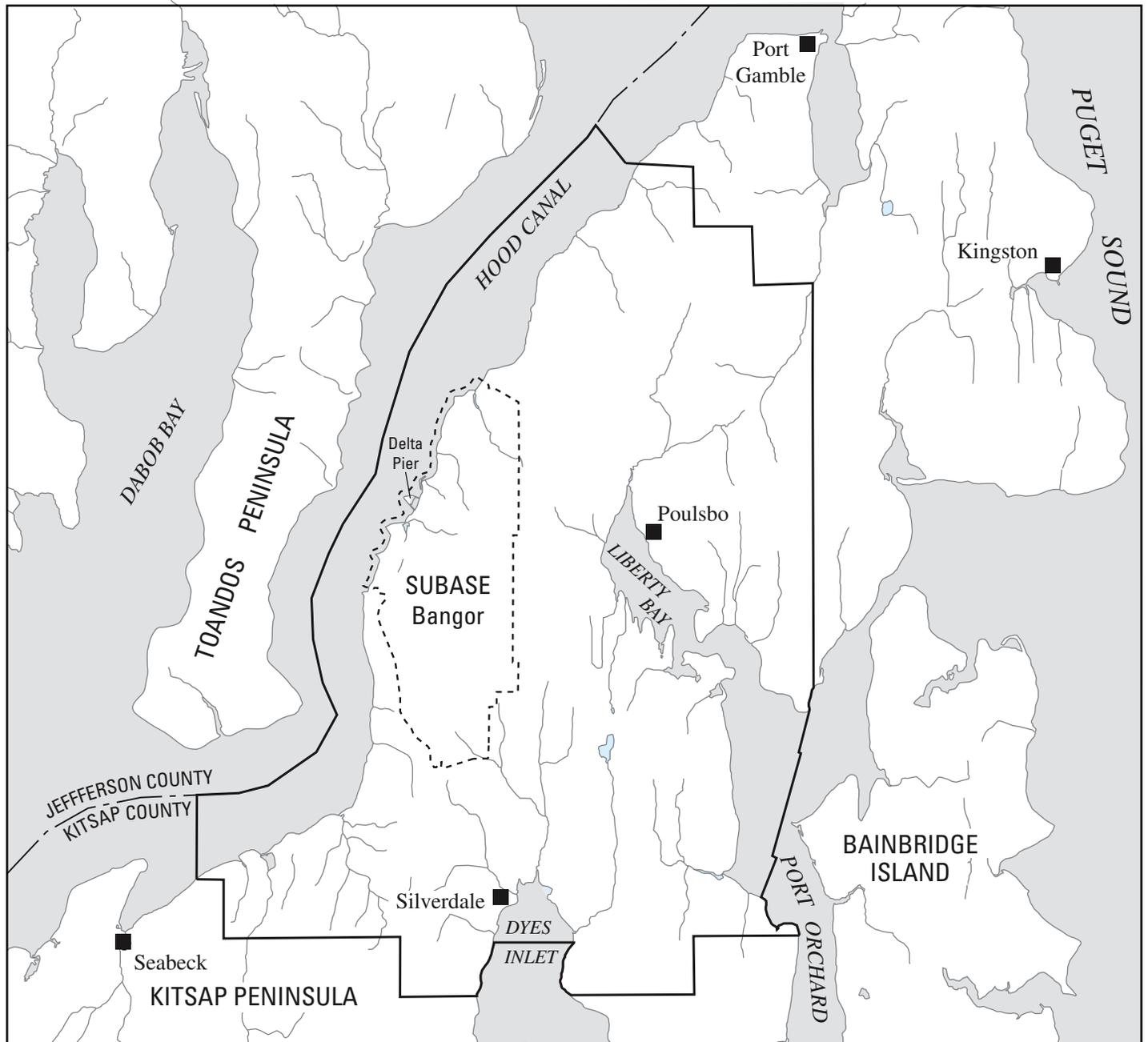# Estimates of Residence Time and Related Variations in Quality of Ground Water Beneath Submarine Base Bangor and Vicinity, Kitsap County, Washington

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Estimates of Residence Time and Related Variations in Quality of Ground Water Beneath Submarine Base Bangor and Vicinity, Kitsap County, Washington

By S.E. Cox

U.S. DEPARTMENT OF THE INTERIOR

GALE A. NORTON, *Secretary*

U.S. GEOLOGICAL SURVEY

Charles G. Groat, *Director*

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CONVERSION FACTORS AND DATUMS

CONVERSION FACTORS

Multiply	By	To obtain
foot (ft)	0.3048	meter
gallon per minute (gal/min)	0.06309	liter per second
inch (in.)	2.54	centimeter
inch (in.)	25.4	millimeter
inch per year (in/yr)	2.54	centimeters per year
mile (mi)	1.609	kilometer
picoCurie per liter (pCi/L)	0.037	becquerel per liter
square mile (mi^2)	2.590	square kilometer
tritium unit (TU)	3.2	picoCurie per liter

Temperature in degrees Fahrenheit (°F) may be converted to degrees Celsius (°C) as follows:

$$°C=(°F-32)/1.8.$$

Temperature in degrees Celsius (°C) may be converted to degrees Fahrenheit (°F) as follows:

$$°F=1.8 \times °C+32.$$

Specific conductance is given in microsiemens per centimeter at 25 degrees Celsius (μS/cm at 25 °C).

Concentrations: Chemical constituents in water are given either in milligrams per liter (mg/L) or micrograms per liter (μg/L). Chlorofluorocarbon concentrations are given in picograms per kilogram of water (pg/kg). Concentrations of tritium are given in tritium units (TU). Concentrations of carbon-14 are expressed as percent modern carbon (pmc). Isotopic ratios are expressed as per mil, or parts per thousand.

Molar concentration units (millimoles or micromoles) are used in discussion of chemical reaction stoichiometry. Molar concentrations are computed as mass of solute divided by formula mass weight of solute per liter of solution.

DATUMS

Vertical coordinate information was referenced to the National Geodetic Vertical Datum of 1929 (NGVD29).

Horizontal coordinate information was referenced to the North American Datum of 1927 (NAD27).

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ABSTRACT

Estimates of residence time of ground water beneath Submarine Base Bangor and vicinity ranged from less than 50 to 4,550 years before present, based on analysis of the environmental tracers tritium, chlorofluorocarbons (CFCs), and carbon-14 (^{14}C), in 33 ground-water samples collected from wells tapping the ground-water system. The concentrations of multiple environmental tracers tritium, CFCs, and ^{14}C were used to classify ground water as modern (recharged after 1953), pre-modern (recharged prior to 1953), or indeterminate. Estimates of the residence time of pre-modern ground water were based on evaluation of ^{14}C of dissolved inorganic carbon present in ground water using geochemical mass-transfer modeling to account for the interactions of the carbon in ground water with carbon of the aquifer sediments.

Ground-water samples were obtained from two extensive aquifers and from permeable interbeds within the thick confining unit separating the sampled aquifers. Estimates of ground-water residence time for all ground-water samples from the shallow aquifer were less than 45 years and were classified as modern. Estimates of the residence time of ground water in the permeable interbeds within the confining unit ranged from modern to 4,200 years and varied spatially. Near the recharge area, residence times in the permeable interbeds typically were less than 800 years, whereas near the discharge area residence times were in excess of several thousand years. In the deeper aquifers, estimates of ground-water residence times typically were several thousand years but ranged from modern to 4,550 years. These estimates of ground-water residence time based on ^{14}C were often larger than estimates of ground-water residence time developed by particle-

tracking analysis using a ground-water flow model. There were large uncertainties—on the order of 1,000-2,000 years—in the estimates based on ^{14}C. Modern ground-water tracers found in some samples from large-capacity production wells screened in the deeper aquifer may be the result of preferential ground-water pathways or induced downward flow caused by pumping stress.

Spatial variations in water quality were used to develop a conceptual model of chemical evolution of ground water. Stable isotope ratios of deuterium and oxygen-18 in the 33 ground-water samples were similar, indicating similar climatic conditions and source of precipitation recharge for all of the sampled ground water. Oxidation of organic matter and mineral dissolution increased the concentrations of dissolved inorganic carbon and common ions in downgradient ground waters. However, the largest concentrations were not found near areas of ground-water discharge, but at intermediate locations where organic carbon concentrations were greatest. Dissolved methane, derived from microbial methanogenesis, was present in some ground waters. Methanogenesis resulted in substantial alteration of the carbon isotopic composition of ground water.

The NETPATH geochemical model code was used to model mass-transfers of carbon affecting the ^{14}C estimate of ground-water residence time. Carbon sources in ground water include dispersed particulate organic matter present in the confining unit separating the two aquifers and methane present in some ground water. Carbonate minerals were not observed in the lithologic material of the ground-water system but may be present, because they have been found in the bedrock of stream drainages that contribute sediment to the study area.

INTRODUCTION

Naval Submarine Base Bangor (SUBASE Bangor) is a U.S. Navy installation of approximately 11 mi^2 (square miles) that has been in operation since 1944. It is located along Hood Canal in Kitsap County, Washington (fig. 1). Ground water from a multi-layered aquifer system is the principal source of water for SUBASE Bangor and the surrounding area. A steady increase in the residential population of the area has resulted in increased awareness of the potential for contamination and increased pressure for the development of additional ground-water supplies. Historical military-industrial activities at SUBASE Bangor have resulted in contamination of shallow ground water and soil at many locations within the base area. Contaminants include ordnance compounds, metals, chlorinated organics compounds, petroleum hydrocarbons, and pesticides. As of 2000, remediation and cleanup have taken place or are in progress at all of the previously identified contaminated locations at SUBASE Bangor.

The U.S. Navy Engineering Field Activity Northwest (EFANW), Naval Facilities Engineering Command, recognized that understanding the regional ground-water flow system beneath SUBASE Bangor was important to understanding the potential movement of contaminants within the multi-layered ground-water system. In 1993, EFANW, in cooperation with the U.S. Geological Survey (USGS), began an investigation of the hydrology and water quality of SUBASE Bangor and its vicinity. Reports have been published on four aspects of the investigation: hydrogeology of the study area (Kahle, 1998); ambient quality of ground water (Greene, 1997); recharge to the shallow aquifer (Bidlake and Payne, 2001); and a three-dimensional numerical simulation of ground-water flow (van Heeswijk and Smith, 2002). The study in this report focused on estimating the residence time of water at various locations in the ground-water system of the study area.

Purpose and Scope

The purpose of this report is to estimate the residence time of ground water at locations within the regional ground-water system beneath SUBASE Bangor and vicinity. The estimates of ground-water residence time will provide information on the duration of ground-water flow between the points of recharge and discharge. The estimates of ground-water residence times were based on the concentration of selected environmental tracers measured in ground-water samples obtained from existing wells that were located along inferred flow paths between areas of ground-water recharge and discharge. The environmental tracers used to estimate ground-water residences times were atmospheric components that enter ground water as dissolved gases at the point of recharge. To estimate ground-water residence times, the concentrations of these dissolved constituents were measured in the ground-water samples and related to historical atmospheric concentrations indicative of the time of recharge. The results of this study were used as one of several parameters to calibrate a numerical simulation model of ground-water flow within the study area (van Heeswijk and Smith, 2002).

Water samples were collected from 33 wells in the area to determine the concentrations of a combination of environmental tracers, including carbon-14 (^{14}C), tritium, and chlorofluorocarbons (CFCs), as well as the concentrations of major ions, isotopic ratios of deuterium, oxygen-18 (^{18}O), and carbon-13 (^{13}C), and the physical properties temperature, oxidation-reduction potential (ORP), and specific conductance. Results of the analyses of environmental tracers and isotope ratios were used to classify the age of ground water at various locations as pre-modern, modern, or indeterminate. Results of the analysis of major ions in water samples were used to develop a conceptual model of water-quality variations along ground-water flow paths, which was then used to guide and constrain the geochemical model used to interpret the ^{14}C data.

Figure 1. Location of the study area, SUBASE Bangor and vicinity, Kitsap County, Washington.
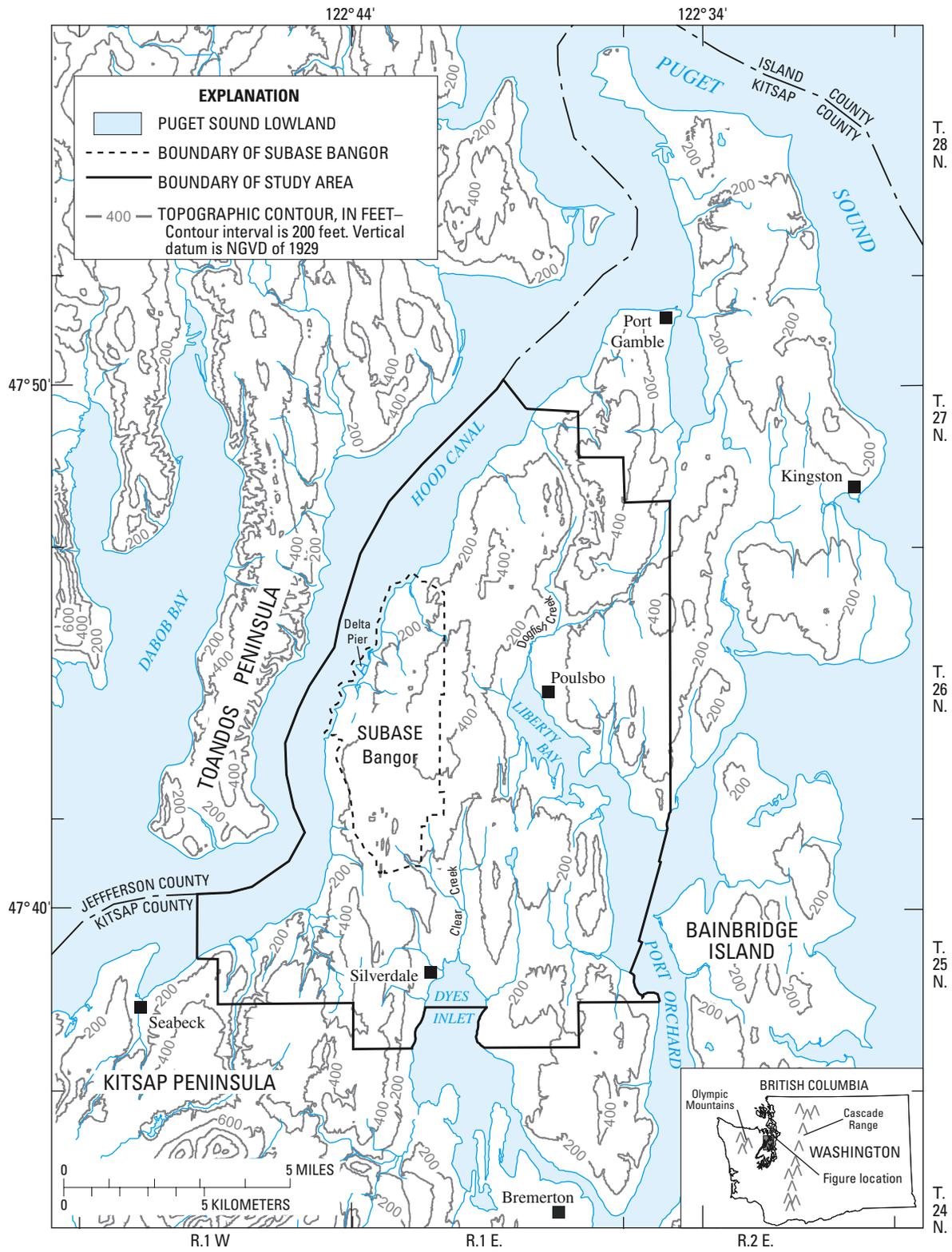
Base map modified from
U.S. Geological Survey digital data, 1:24,000
Universal Transverse Mercator projection,
Zone 10

Description of Study Area

The project study area includes SUBASE Bangor (about 11 mi^2) and roughly 65 mi^2 of the adjacent area on the western half of the Kitsap Peninsula ([fig. 1](#)). The study area boundaries were selected to include hydrologic boundaries to be used in the numerical simulation of ground-water flow. The Kitsap Peninsula extends into Puget Sound, with Hood Canal to the west and Puget Sound to the north and east. Hood Canal, along which SUBASE Bangor is located, is a U-shaped fjord with steep flanks rising abruptly from the coast to altitudes of 200 ft above sea level or more. Inland, slopes are moderate and many areas are nearly flat. Maximum altitudes in the interior of the peninsula are less than 500 ft. Streams flow only a short distance from the interior of the peninsula before reaching the marine waters of Hood Canal or Puget Sound. About 13 percent of the area is built up as urban or military development and about 47 percent of the study area is covered with coniferous and deciduous forests. The remaining 40 percent of the study area is covered by non-forest vegetation including agricultural lands and natural vegetative shrub cover. The geology of the study area is characterized by glacial and nonglacial sedimentary deposits forming alternating beds of more permeable and less permeable sediments that range in total thickness from 600 to 1,800 ft (Jones, 1996). Glacial deposits include primarily tills, outwash fluvial sands and gravels, and lacustrine silt-clay deposits. Non-glacial deposits include floodplain deposits that often include peat and buried soils.

Compared to the rest of the Puget Sound region, the Kitsap Peninsula is largely undeveloped. The population of the study area is concentrated in the two medium-sized towns of Silverdale and Poulsbo, with 1990 populations of 7,660 and 4,848, respectively (U.S. Bureau of the Census, 1992). The resident population of SUBASE Bangor was 2,830 in 1993. Outside these areas, the countryside is rural and semi-rural, with many homes obtaining potable water from private wells instead of public-supply systems. From 1970 to 1990, the population of Silverdale and Poulsbo increased by about 150 percent. The increase in population is expected to continue, with growth from about 39,000 inhabitants in 1990 to about 76,000 in 2020 (Puget Sound Council of Governments, 1988; U.S. Bureau of the Census, 1992). The population of SUBASE Bangor has been projected to increase to 6,372 in 2012, as additional residential housing is constructed on base (Parametrix, 1994).

The study area has a temperate maritime climate. Mean annual precipitation ranges from approximately 30 in/yr (inches per year) in the northeastern part of the study area to approximately 60 in/yr in the southwestern part (Kitsap County Ground Water Advisory Committee and others, 1991). Precipitation amounts are controlled in large part by the Olympic Mountains to the west and the Cascade Range to the east, which impede the flow of humid air masses generated over the Pacific Ocean. In general, precipitation reaches a minimum during mid-summer (July through August) and a maximum during the late fall and early winter (November through January). The mean monthly temperature in the study area ranges from approximately 39°F (degrees Fahrenheit) in January to 64°F in July and August (Owenby and Ezell, 1992). At times, winter temperatures are sufficiently cold that several inches of snow may accumulate; however, snow accumulation generally is not significant.

Precipitation is the source of all surface and ground water in the study area. Short streams flow from the interior of the peninsula to the Hood Canal or the larger drainages of Clear and Dogfish Creeks, which drain to Puget Sound. Most streams flow year-round and are fed by springs, aquifer discharge, and surface runoff following storms. Where cliffs are present along the coastline, springs and seeps discharge water directly onto the beach and into Puget Sound. The fraction of the annual amount of precipitation that infiltrates through the soil to reach the ground-water system is recharge. The amount of recharge varies spatially because of differences in the amount of precipitation, soil and geological characteristics, land-use practices, and vegetation. Estimated long-term average recharge to the shallow aquifers of the study area typically range from 8 to 10 in/yr, but can range from 0 to about 21 in/yr (Bidlake and Payne, 2001). Deeper aquifers are recharged by ground water from the overlying aquifers. Roughly 40 percent of recharge to the shallow aquifer passes through to recharge deeper aquifers (van Heeswijk and Smith, 2002). The direction of ground-water flow on the peninsula is primarily toward coastal areas, particularly in the deeper aquifers, however, in shallow parts of the flow system, ground-water flow is toward Hood Canal and the interior drainages of Dogfish and Clear Creeks. Most ground water within the study area discharges to Hood Canal.

Previous regional studies of the ground-water quality in Kitsap County characterized the quality of ground water as good and suitable for most uses, although concentrations of iron and manganese were found to exceed secondary guidelines in some wells (Garling and others, 1965). Van Denburgh (1965) found that ground water from the Sea-level aquifer (referred to as Salmon Springs in his report) contained distinctly larger concentrations of potassium and orthophosphate than those found in near-surface glacial sediments. Concentrations of silica also generally were larger in ground water from the deeper sediments. The larger concentrations were attributed to longer periods of contact with the host rock material. More recently, as part of this study, Greene (1997) sampled 136 wells to determine ambient background concentrations of numerous inorganic and organic constituents; the median concentrations of selected constituents from that study are listed in .

Table 1. Median values of physical properties and concentrations of water-quality constituents in ground water from five hydrogeologic units, SUBASE Bangor and vicinity, Kitsap County, Washington

[**Median concentrations** are from Greene (1997) and are given in milligrams per liter, unless otherwise noted. **Hydrogeologic units:** Number of samples from each unit is noted in parentheses. Qvt, Vashon till confining unit; Qva, Vashon aquifer; QC1pi, permeable interbeds within Upper confining unit; QC1, Upper confining unit; QA1, Sea-level aquifer. **Abbreviations:** °C, degrees Celsius; µS/cm, microsiemens per centimeter at 25 degrees C; mg/L, milligrams per liter; –, no data]

Physical properties and water-quality constituents	Median values and concentrations by hydrogeologic unit				
	Qvt (3)	Qva (54)	QC1pi (34)	QC1 (16)	QA1 (29)
Temperature (°C)	–	10.8	9.6	11.5	–
Specific conductance, field (µS/cm)	172	151	158	161	157
pH, field (units)	6.7	7.6	7.9	8.0	8.1
Dissolved oxygen	5.6	6.3	.4	.6	1.2
Alkalinity, as $CaCO_3$	45	190	75	230	–
Hardness, as $CaCO_3$	85	68	130	26	–
Calcium, dissolved	14	11	14	15	15
Magnesium, dissolved	10	8.0	7.8	7.8	6.8
Sodium, dissolved	5.0	4.6	5.2	6.10	5.5
Potassium, dissolved	.6	.8	1.3	1.1	1.4
Bicarbonate, dissolved	97	80	92	94	96
Sulfate, dissolved	4.2	3.9	1.6	3.2	1.8
Chloride, dissolved	2.8	2.5	2.1	2.3	1.9
Fluoride, dissolved	<.1	<.1	<.1	<.1	<.1
Silica, dissolved	29	28	30	30	26
Iron, dissolved	.029	.010	.038	.042	.018
Manganese, dissolved	<.001	.001	.038	.043	.014
Ammonium-N, dissolved	.02	<.15	.04	.04	.08
Nitrite-N, dissolved	<.01	<.01	<.01	<.01	<.01
Nitrate + nitrite-N, dissolved	.73	.19	<.05	<.05	<.05
Phosphate-PO_4, dissolved	.02	.04	.08	.09	.12
Aluminum, dissolved	<.01	<.01	<.01	<.01	<.01
Boron, dissolved	.008	.005	.008	.005	.010
Bromide, dissolved	.02	.01	.01	.02	.02
Lithium, dissolved	<.004	<.004	<.004	<.004	<.004
Strontium, dissolved	.093	.053	.056	.070	.061

Volatile organic compounds, indicative of anthropogenic land-use activities, were detected in two of the wells sampled by Greene (1997). Localized studies related to cleanup of contaminated sites on SUBASE Bangor have documented the presence of anthropogenic contaminants in the shallow aquifer at some sites. Direct information regarding estimates of ground-water residence time was not found in previous water-quality investigations; however, the presence in ground water of anthropogenic contaminants related to land-use activities of the past 50 years indicates that the age of shallow ground water is less than 50 years.

Well-Numbering System

In Washington, wells are assigned local well identification numbers based on their location with respect to the Willamette Base Line and Meridian using the Public Land Survey system of township, range, section, and 40-acre tract (fig. 2). Well number 26N/01E-12Q01 indicates that the well is in the southwest quarter of the southeast quarter of section 12 in township 26 North (N) and range 1 East (E) of the Willamette Base Line and Meridian. The two-digit sequence number (01) following the letter designation indicates that this was the first well inventoried by USGS personal in that 40-acre parcel; in situations where a well has been deepened, a "D" is added at the end of the local well number.
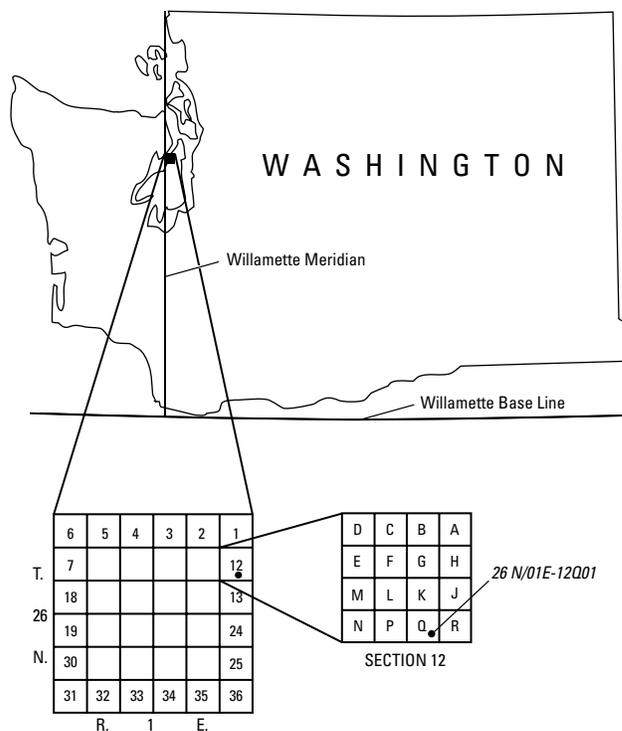
Figure 2. Well-numbering system used in Washington.

Acknowledgments

BACKGROUND CONCEPTS ON AGE-DATING OF GROUND WATER

Nearly all ground-water recharge is derived from atmospheric precipitation containing dilute concentrations of many constituents that are present in the atmosphere. Several atmospherically derived ground-water constituents, including carbon dioxide, CFCs, and tritium, can function as a record of ground-water recharge. Each of these indicators can be used to assess ground-water age if the initial concentration is known and the chemical reactions and physical processes affecting its concentration within the ground-water system are understood and can be predicted. The residence time of ground water, also referred to as the age of ground water, typically implies the elapsed time between ground-water recharge and the time when the ground-water sample was collected. More specifically, ground-water residence time begins at the point when recharging ground waters are isolated from the earth's atmosphere. In relatively shallow ground-water systems, this typically occurs at or just below the water table.

The atmospheric components used as environmental tracers to estimate the residence time of ground water in this study were ^{14}C (contained in carbon dioxide), tritium, and a suite of three CFCs. Tritium and CFCs are present in ground water largely as a result of human activity during the last 40 to 50 years. Therefore, the presence of tritium and CFCs in ground water indicates that recharge had occurred within the last half-century, and these ground waters are referred to young or "modern." Estimates of ground-water ages of older, or "pre-modern" ground waters can be made using ^{14}C. Under optimal conditions, ^{14}C determinations can provide estimates of ground-water residence times roughly between 500 and 35,000 years. Analytical procedures are not routinely available to quantify tracers that could be used to estimate ground-water ages in the range of 50 to 500 years (Plummer and others, 1993).

All methods that use environmental tracers to estimate the elapsed time between ground-water recharge and sample collection involve a number of underlying assumptions regarding both the regional or global input of the environmental tracers into the ground-water system and the physical, chemical, and biological processes that act on the tracer as it moves through the ground-water system. Estimates of ground-water age typically require extensive interpretation of the tracer-concentration data, and the resulting estimated residence time should not be considered as a true age but more appropriately as a "model age" (Plummer and others, 1993) or as a "ground-water mean residence time" (Clark and Fritz, 1997). In this report, "model age" is used to indicate the estimate of the average elapsed time between ground-water recharge and the time when the ground-water sample was collected, and is based on the interpretation of the tracer concentrations and ancillary data.

Tritium

Tritium, the radioactive isotope (^{3}H) of the element hydrogen, is an excellent tracer of ground water because it is incorporated into water molecules of rainfall that recharges ground water, and its movement within the ground-water system is not restricted by chemical or physical processes. Tritium is continually produced in small quantities in the upper atmosphere by the interaction of cosmic rays with gaseous nitrogen. However, its importance as a ground-water tracer is due largely to the tremendous amounts of tritium that were produced in the atmosphere as a result of the above-ground testing of thermonuclear weapons between 1952 and 1980. The cessation of large-scale atmospheric testing of nuclear weapons in 1962 was followed by a spike in atmospheric tritium concentration in 1963 that was several hundred times larger than natural background concentrations. The half-life of tritium is 12.4 years, and elevated tritium concentrations have persisted in the atmosphere for many decades following the cessation of atmospheric nuclear weapons testing. The atmospheric concentration of tritium in western Washington was estimated to range from 3 to 5 TU (tritium units) prior to nuclear weapons testing (Thatcher, 1962). Radioactive decay over a period of 40 years would have reduced that concentration to about 0.3 to 0.5 TU.

Thus, the presence of tritium in ground-water samples at concentrations greater than 1 TU indicates that the ground water was recharged after the start of atmospheric testing of thermonuclear weapons. Atmospheric tritium from weapons testing (often referred to as bomb tritium) continues to decay, and tritium concentrations in present-day rainfall are still distinguishable from tritium concentrations in ground water recharged prior to the period of bomb testing.

The absence of tritium in ground-water samples clearly indicates recharge prior to the period of bomb testing. Such ground water is designated either as pre-modern or older ground water. Conversely, ground water containing tritium concentrations greater than 1 TU is considered modern or young. Ground water containing tritium concentrations between the detection limit and 1 TU may be considered to have been recharged prior to the bomb testing period (Mazor, 1991) or is predominantly older water that may have mixed with a smaller fraction of modern ground water recharged after 1953 (Clark and Fritz, 1997), and is referred to as indeterminate in this report. Mixing can take place either in the aquifer along converging ground-water flow paths, within the well bore, as an artifact of well construction, or by diffusion from adjacent confining units (Goode, 1996).

Chlorofluorocarbons

CFCs are a class of synthetic organic compounds that are thought to be entirely of anthropogenic origin, and when present in ground water can be used to estimate ground-water residence time (Plummer and Busenberg, 2000). Production of CFCs began in the 1930s and their primary use was as refrigerants, but they also have a variety of other uses. Ultimately, CFCs are released to the atmosphere, where the concentrations of chlorotrifluoromethane (CFC-11), dichlorodifluoromethane (CFC-12), and trichloro-trifluoroethane (CFC-113) can be detected. A record of the atmospheric concentrations of these CFCs has been developed from measurements made at Niwot Ridge, Colo., and from CFC production records.

Concentrations of CFC-11 and CFC-113 in the atmosphere have increased steadily from their earliest detections until peak concentrations were reached in 1993 and 1994, respectively, whereas concentrations of CFC-12 were still increasing in 1994 and 1995 when the samples were collected for this study. CFCs in the atmosphere can dissolve into falling precipitation or into soil moisture in the unsaturated zone and thereby impart a signal in the ground water of the time of recharge. The measured concentrations of CFCs in a ground-water sample can be used to calculate the corresponding equilibrium vapor concentrations (using Henry's Law) that can be compared to historical atmospheric concentrations to determine the recharge date. The concentration of individual CFCs and the mixture of specific CFCs thus can be used to date some ground waters that recharged after 1940. Concentrations of CFCs in ground water that are larger than those corresponding to maximum atmospheric concentrations indicate additional CFC sources, such as landfills, abandoned refrigerators, and air conditioners leaking directly into the aquifer, as well as recharge from sanitary septic systems. Although these larger concentrations exceeded the range from which reliable estimates of ground-water residence time can be derived, they generally are orders of magnitude smaller than concentrations cited as drinking-water or aquatic-health guidelines.

Carbon-14

^{14}C is the radioactive isotope of carbon, and its concentration in ground water, in the form of dissolved inorganic carbon, has been used routinely since 1970 to estimate the age of ground water. ^{14}C is produced continuously in the upper atmosphere by the interactions of nitrogen and cosmic rays. ^{14}C-carbon dioxide ($^{14}CO_2$) can enter ground water while recharge water is in contact with the atmosphere, occurring mostly in the unsaturated zone where the gaseous concentration of $^{14}CO_2$ is substantially increased by root respiration and microbial oxidation of organic matter.

Once recharge water enters the aquifer at the water table and becomes isolated from the atmosphere as a result of downward flow, ^{14}C in ground water will diminish due to radioactive decay at a rate of one-half of the remaining carbon every 5,730 years. ^{14}C age estimates generally are expressed in terms of years before present (bp), whereas the concentration of ^{14}C in ground water is expressed as percent modern carbon (pmc). By convention, the concentration of "modern carbon" is defined as 13.56 disintegrations per minute per gram carbon and is assigned an initial date of 1950 (Mook, 1980; Kalin, 2000). The selection of 1950 is due in part to the injection of large concentrations of ^{14}C into the atmosphere following the atmospheric testing of nuclear weapons. Concentrations of ^{14}C in ground water recharged after the advent of nuclear weapons testing in 1950 often are larger than 100 pmc and are often referred to as containing bomb carbon.

A ^{14}C ground-water age is computed from the half-life age equation derived from the radiometric decay equation written in the form:

$$T = -5,730/\ln2 * \ln\{A_t{}^{14}C/A_o{}^{14}C\}, \qquad (1)$$

where

    5,730 = the half-life of ^{14}C, in years,
    $A_t{}^{14}C$ = the measured ^{14}C activity in the downgradient well, and
    $A_o{}^{14}C$ = the ^{14}C activity in the upgradient well or recharge area.

Direct application of this equation involves two key assumptions that may in fact not be true, particularly for recently recharged ground water. The first assumption is that $A_o{}^{14}C$ is known and has remained constant. The second is that carbon does not enter or leave ground water other than by radioactive decay. Because both of these assumptions often are not accurate, the difficulty in applying radio-carbon dating to ground water is in determining the extent to which physical, chemical, and biological processes have affected the ^{14}C content of ground water in the upgradient (recharge) area and along the ground-water flow path to the sampled well.

To provide better approximations of ground-water residence time based on ^{14}C concentrations of dissolved inorganic carbon, a variety of models have been developed to account for geochemical processes that affect the ^{14}C concentrations in ground water. Typically, these models estimate the mass of carbonate and organic carbon that accumulates in ground water as it moves through the hydrogeologic system. Essential to all of these models are the assumptions relating to the temporal consistency of the ^{14}C activity at the time of recharge and to the origin of the carbon added to or removed from ground water along the flow path. Examples of such models include those of Ingerson and Pearson (1964), Tamers (1967), Mook (1976), Wigley and others (1978, 1979), and Fontes and Garnier (1979). In this study, the stable isotope of carbon, carbon-13 (^{13}C), and ancillary chemical data from the ground-water samples are used to estimate the extent to which chemical and microbial processes have affected the ^{14}C concentration through the use of NETPATH, an inverse geochemical mass-transfer modeling approach developed by Plummer and others (1991, 1996).

HYDROGEOLOGIC SYSTEM

Ground water beneath the study area is found in a series of aquifers composed of permeable sand and gravel layers separated by layers of less permeable deposits of silt, sand, and clay (fig. 3). The movement of ground water is controlled by recharge, topographic geometry, and the hydraulic properties of the sedimentary material. Generally, ground water moves from areas of topographic highs to topographic lows along the coast or stream drainages. The direction of ground-water flow may be upward in ground-water discharge zones along the coast and stream drainages. Typically, the direction of ground-water flow in permeable aquifer layers approaches horizontal, whereas flow in the semi-confining and confining units is closer to vertical. Estimates of long-term average ground-water recharge in most of the study area range from 8 to 10 in/yr (Bidlake and Payne, 2001). The unsaturated zone between land surface and the water table is typically 60 to 80 ft in the higher altitudes near the central part of the study area and much smaller near areas of ground-water discharge.

WEST   **EAST**

Hood Canal SUBASE Bangor Qvt Qva QC1pi QC1 QC1pi *Clear Creek Drainage* Qvt Qva Qvr SEA LEVEL QA1 QC1 QC QA2 QC QC QC QU Br

NOT TO SCALE

EXPLANATION

PERMEABLE UNITS

Qvr   Surficial aquifer

Qva   Vashon aquifer

QC1pi   Permeable interbeds within Upper confining unit

QA1   Sea-level aquifer

QA2   Deep aquifer

LESS PERMEABLE UNITS

Qvt   Vashon till confining unit

QC1   Upper confining unit

QC   Undifferentiated confining units

QU   Undifferentiated deposits

Br   Bedrock

- - - →   CONCEPTUALIZED GROUND-WATER FLOW PATH

Figure 3. Conceptual model of hydrogeologic units and ground-water flow in the vicinity of SUBASE Bangor, Kitsap County, Washington. (Modified from Kahle, 1998.)

The hydrogeologic framework of the study area is dominated by a thick accumulation of glacial and non-glacial sediments that have been deposited over the last several hundred thousand years, resulting in a sequence of alternating coarse (permeable) and fine-grained (less permeable) deposits that partially fill the regional north-south structural depression referred to as the Puget Sound Lowland. In the area of SUBASE Bangor, the thickness of these sediments is typically greater than 1,000 ft (Jones, 1996). Except for the sediments deposited by the last receding glacier, sediments in the study area were overridden and compacted by more than 3,000 ft of ice during the last glacial maximum (Booth and Goldstein, 1994).

During successive periods of continental glaciation, glacial sediments were transported to and deposited within the Puget Sound Lowland by southward-moving glaciers and outwash streams. The sedimentary material originated largely from the metamorphic and granitic terrains of the North Cascades and British Columbia Coast Mountains.

These glacial deposits generally consist of sands and gravels, lacustrine silts and clay, and till. Outwash sand-and-gravel deposits typically are permeable and make excellent aquifers. In contrast, glacial tills and lacustrine deposits of silts and clay are much less permeable and act as confining units. Grain-size distributions in these glacial deposits are often heterogeneous, creating stratigraphic irregularities that affect the flow of ground water. Lenses of fine-grained sediments present in the outwash sands and gravels often reduce or divert the flow of ground water. Conversely, within the fine-grained sediment of the confining units are lenses of coarse-grained sediments of sufficient extent and permeability to provide quantities of ground water for domestic wells.

Non-glacial sediments were deposited in the Puget Sound Lowland during periods when continental glaciers were absent. These sediments consist largely of low-energy floodplain deposits, but in some areas may also contain sands and gravels characteristic of alluvial fans. The non-glacial sediments originated from streams and rivers draining the adjacent Olympic and Cascade uplands, which border the Puget Sound Lowland on the west and east. Near the study area, patterns of sedimentary cross bedding in the non-glacial sediments indicate a southwest-to-northeast pattern, suggesting flow from the Olympic Mountains (Borden, 1998). The sedimentary material from the Olympic Mountains was characterized as basalt, graywacke, and argillite, whereas andesite and metavolcanics were more characteristic of Cascade uplands (Borden and Troost, 2001). Alpine glaciers originating in the Cascade and Olympic Mountains are thought to have been most active and extended farthest into the Puget Sound Lowland in the period just prior to the arrival of each successive continental glacier.

The description of stratigraphy of the upper half of this complex sequence of sediments in the Bangor area has been refined by the studies by Noble (1989 and 1990), Becker (1995a), Borden (1998), and Kahle (1998). The stratigraphy and hydrogeologic units used in this report (fig. 3) are those described by Kahle (1998). The additional lithologic and mineralogic descriptions provided below are useful in understanding the geochemical mass-transfer models.

Vashon Till Confining Unit

The uppermost subglacial lodgement till is referred to as the Vashon till confining unit (Qvt), and its surficial exposure covers about 71 percent of the study area (Kahle, 1998). Recessional outwash (Qvr, called the Shallow aquifer by Kahle) covers an additional 14 percent of the area, some of which may also overlay the Qvt. The Vashon till is a pebbly and silty sand with roughly 10 to 20 percent clay-sized fragments (Vanderwal, 1985). Although the permeability of this till is typically very low (10^{-7} to 10^{-5} centimeters per second), more-permeable lenses are interspersed within the till and can yield small quantities of water. Thickness of the till ranges from 0 to greater than 200 ft, but is typically 10 to 100 ft.

Petrologic examination of rock clasts obtained from the Vashon till at sites throughout the Puget Sound Lowland indicate that nearly 60 percent of the clasts found in the till were of metamorphic origin, and the remaining clast were of granitic, volcanic, or clastic origins (Vanderwal, 1985). Clasts from sites near the Bangor study area were classified as 63 percent metamorphic, 14 percent clastic sedimentary rocks, 12 percent granitic, 7 percent andesitic volcanics, 3 percent chert, and 2 percent basaltic volcanics (Vanderwal, 1985). Mineralogical analyses of samples of the Vashon till collected from the eastern part of the Puget Sound Lowland by Mullineaux (1970) and Mullineaux and others (1964) show that carbonate minerals were rarely present in these samples. The silt-size fraction typically was 50 percent quartz and 40 percent plagioclase with a trace of chlorite. The clay-size fraction of Vashon till included 40 to 50 percent montmorillonite; 20 percent chlorite; 10 percent quartz; and 10 percent plagioclase. The heavy minerals identified in the 1/16 to 1/4 mm (millimeter) size fraction of these samples included pyroxene, amphibole (chiefly hornblende), garnet, and iron-magnesium minerals and rock fragments. In one sample, siderite (an iron-carbonate mineral) was measured as less than 5 percent of the heavy mineral fraction, or less than 1 percent of the total sample. There was no other mention of carbonate-bearing minerals except in the much deeper and older till deposit not encountered in the Bangor study area.

Vashon Aquifer

The Vashon aquifer (Qva) is composed of advance glacial outwash, which generally is a well-sorted deposit of gravelly sands with occasional lenses of fine-grained material. This unit occurs widely in the study area and typically is 10 to 150 ft thick, but can be as much as 250 ft thick. Within the study area, sediment grain size in the Vashon aquifer changes from fine to medium sands in the stratigraphically lower portions to coarse sands and gravels in the stratigraphically higher portions where foreset bedding is common (Deeter, 1979). The advance outwash is exposed at the surface in 11 percent of the study area where the Vashon till is absent. These windows through the till to the Vashon aquifer typically are along drainages or near-shore areas, although several are in the upland recharge areas (Kahle, 1998).

The lithologic and mineral composition of the Vashon outwash deposits has not been well documented. Based on the similarity in sediment source areas, the composition of the Vashon till and outwash are expected to be similar. Major minerals identified in Vashon outwash, obtained from a surface-exposure sample, include quartz and plagioclase, with minor fragments of tremolite, actinolite, microcline, epidote, and chlorite. Borden (1998) reported that the sands of the Vashon outwash are composed predominantly of quartz, with minor amounts of muscovite, and that lithic fragments make up less than 30 percent of the grains. Likewise, gravel clasts are a mixture of felsic intrusive, dacite, andesite, and basalt and meta-volcanic sediments all of which were indicative of a northern source area. Schlichte (1968) reported that the mineralogical composition of a soil developed on the outwash was composed of roughly 44 percent plagioclase, 26 percent potassium feldspar, 16 percent quartz, and 14 percent heavy minerals, which were mostly pyroxenes and amphiboles. Mullineaux (1967) reported that quartz and feldspars composed 90 percent of two samples of Vashon drift silt; and that the heavy-mineral content was primarily altered iron-magnesium minerals and rock fragments (35-50 percent), pyroxene (22-45 percent), amphibole (hornblende; 5-25 percent), garnet (2-10 percent), and opaques (5-15 percent). Neither calcite nor siderite was found in any of 13 samples, nor was there reference to any other carbonate minerals.

Upper Confining Unit and Interbeds

A diverse group of generally low-permeability sediments beneath the Vashon glacial aquifer sediments separates the Vashon aquifer from the lower aquifers. Within the study area, Kahle (1998) classified this diverse group of sediments as a single hydrogeologic unit with the designation of QC1. The QC1 hydrogeologic unit is extensive, covering the entire study area. Thickness of the QC1 generally is between 100 and 300 ft, but may exceed 500 ft. The geologic stratigraphy within QC1 is complex and includes the Kitsap unit as described by Garling and others (1965), the Kitsap and Whidbey units as described by Deeter (1979), and the Lawton clay of Mullineaux and others (1964). The QC1 unit also contains discontinuous lenses or interbeds of gravelly material. Where mappable, Kahle (1998) classifies these gravelly deposits within the QC1 as permeable interbeds denoted as QC1pi. As mapped by Kahle (1998), the QC1pi is present in about 25 to 30 percent of the study area and lenses typically are about 30 ft thick but may exceed 100 ft thick (Kahle, 1998). The fine-grained sediments of the QC1 are not very permeable, yielding insufficient water for well construction. However, the interbedded gravels and sands (QC1pi) can be quite permeable and typically are capable of providing usable quantities of ground water for domestic supplies. Previous studies by Noble (1975a; 1975b) and Becker (1995a; 1995b) have designated these permeable interbeds in the southern part of the study area as the Semi-perched aquifer.

The sedimentary composition of the QC1 is diverse, including deposits of glaciolacustrine silts and clay (Lawton clay) and lenses of fluvial gravelly sands. However, most of this unit is composed primarily of several non-glacial floodplain and lacustrine deposits of clayey silt and sands. Within the fine-grained sediments, lenses of silty peat and dispersed organic matter occur. Radiocarbon ages reported by Deeter (1979) from peat or woody material collected within the Bangor study area yield dates between 15,000 and greater than 40,000 years bp. Coarse-grained fluvial sediments within the non-glacial deposits, exposed in the sea cliffs along the coastal margin of SUBASE Bangor, are likely correlative with the permeable interbed hydrostratigraphic unit (QC1pi) described by Kahle (1998; U.S. Geological Survey, oral commun.,

2002). According to Borden (1998), these highly oxidized fluvial sediments are composed predominantly of thick-bedded to massive cross-bedded sands, gravels, and silts. Cross-bedding features indicate that the direction of streamflow during deposition of these sediments was from southwest to northeast. Lithic fragments are composed predominantly of basalt, gabbro, and basaltic sandstone derived from the Crescent Formation of the western Olympic Mountains (Borden, 1998; Noble, 1990). The non-glacial gravels are lithologically very similar to gravels from the Quilecene, Dosewallips, and Duckabush Rivers, which drain the eastern flanks of the Olympic Mountains, further suggesting that the source for sediments in the QC1pi is likely the eastern Olympic Mountains.

The basalts of the Crescent Formation are a thick accumulation of submarine and subaerial basalts with interbedded basaltic breccias, hyaloclastites, and minor fossiliferrous limestone (Glassley, 1973; Tabor and Cady, 1978). The basalts range in age from 45 to 58 million years and are highly altered and, in some instances, mildly metamorphosed to prehnite-pumpellyite and greenschist facies (Glassley, 1973). The mineralogy of basalts of the Crescent Formation was described by Glassley (1973, 1974) as typically composed of plagioclase and pyroxene (augite) with lesser amounts of glass, olivine (present in the basalts), iron-titanium oxides, and apatite. Submarine basalts have less-developed crystalline structure than subaerial basalts because of more rapid cooling, and are more highly altered to secondary minerals. Magnetite and ilmenite present in the original rocks have been extensively altered to hematite and sphene. Basaltic glass is absent, having been altered to secondary minerals such as chlorite, prehnite, and pumpellyite.The mineralogic composition of low-grade alteration of basaltic rocks is typically a mixture of prehnite, pumpellyite, chlorite, actinolite, albite, sphene, and minor amounts of carbonates.

Sea-Level Aquifer and Deep Aquifer

Below the fine-grained non-glacial deposit of the Upper confining unit is a deep aquifer composed of fluvial sands and gravels of basaltic origin, referred to as the Sea-level aquifer (QA1; Noble, 1989; Becker, 1995a, 1995b; and Kahle, 1998). The Sea-level aquifer produces large quantities of water to many municipal wells throughout the study area. Below the Sea-level aquifer is a discontinuous confining unit (QC), the Deep aquifer (QA2), and bedrock. Tertiary bedrock, which underlies the Pleistocene deposits, is estimated to be at depths ranging from about 600 to 1,800 ft below land surface, but typically is greater than 1,000 ft below land surface (Jones, 1996).

STUDY METHODS

The residence time of ground water was estimated at locations within the Bangor aquifer system, including areas of ground-water recharge and discharge. Sampling sites were concentrated in three general areas where conceptualized ground-water flow paths from areas of recharge to areas of discharge could be inferred from hydrologic and lithologic information contained in the reports by van Heeswijk and Smith (2002) and Kahle (1998). Residence times were estimated by comparing the concentrations of environmental tracers observed in ground-water samples from areas of ground-water recharge with those at downgradient locations within the ground-water flow system. The actual flow paths followed by ground water could not be identified, thus it was not possible to specifically identify upgradient and downgradient water chemistries. Variations in the range of common-ion concentrations observed in the Vashon aquifer (Qva) were small (Greene, 1997). Water-quality conditions representing recently recharged ground water were assumed to approximate uniform conditions within the Qva; thus, the median concentration of common ions in the Qva was used as the initial concentration in mass-transfer modeling used to interpret ^{14}C estimates of ground-water residence time observed at downgradient locations. The selection of sampling points along an inferred flow path was constrained by the availability of existing wells, their location, and suitability for the collection of ground-water samples.

Sample Collection and Processing

Ground-water samples were collected from 33 wells in the study area (fig. 4). The location and well-construction information are listed in table 2. Five wells were sampled in June 1994 and the remainder were sampled in March and April 1995. Most of the sampled wells were located near three conceptualized ground-water flow paths (fig. 4). Most samples were collected from the Vashon aquifer (Qva), permeable interbeds in Upper confining unit (QC1pi), and the Sea-level aquifer (QA1). Access to the Sea-level aquifer was limited by the lack of suitable wells tapping this unit in the study area. Eight samples were obtained from the deeper aquifers, however, most of these were from large-capacity production wells. A single sample was obtained from a well screened only in the Deep aquifer (QA2), which occurs below the Sea-level aquifer. A second sample was obtained from a well open to both QA1 and QA2. Wells selected for sampling were primarily domestic wells that typically provide more discrete samples because of their shorter screen interval and lower pumping rates. However, several large municipal wells were sampled where more suitable wells were not available. Submersible pumps were temporarily installed in four test wells on SUBASE Bangor to obtain samples where no other pump system was available.

Sample collection points were located as close to the wellheads as was practical and before passage through pressure tanks or water-treatment systems, following the procedures and guidelines outlined by Classen (1982). Water samples were collected after extensive flushing of the casing and well plumbing. A manifold was used to direct discharge from the well to raw and filtered sampling ports and to a fully enclosed flow chamber that prevented atmospheric contact with the water being analyzed. The flow chamber was used to make field measurements of specific conductance, pH, dissolved-oxygen concentration, and oxidation-reduction potential (ORP). The ORP probe was calibrated using Zobell's solution at 25°C as described by the American Public Health Association and others (1992). The dissolved-oxygen probe was calibrated using air-saturated water corrected to ambient atmospheric pressure. Duplicate dissolved-oxygen samples were collected for analysis by Winkler titration when the stabilized reading from the

dissolved-oxygen probe indicated less than 1 mg/L of dissolved oxygen. Two-point bracketing calibration procedures were used to calibrate pH and specific conductance meters.

Raw and filtered samples were collected from sampling ports connected to the stainless steel manifold. Tritium, deuterium, and ^{18}O samples were collected without head space from raw water discharged to sample bottles equipped with poly-cone seals. Alkalinity in samples, filtered through non-acid cleaned apparatus, was determined by incremental titration. Usually, the alkalinity titration was performed at the sampling sites; however, in some instances this titration was completed upon return to the office. Nutrient samples were collected in amber bottles, chilled to 4°C, and preserved with mercuric chloride. Samples of total organic carbon (TOC) and dissolved organic carbon (DOC) were chilled and stored at 4°C without additional preservatives. Procedures used to process samples for inorganic constituents are described by Wood (1981), with the exception that a 0.1-micrometer filter was used rather than a 0.45-micrometer filter. The 0.1-micrometer filter was used to reduce the potential that colloidal material might augment the dissolved concentrations reported by the laboratory and influence the results of the ensuing geochemical modeling.

Methane samples, devoid of gas bubbles, were collected using a glass syringe and hypodermic needle through a butyl-rubber septum installed in the raw discharge line. Samples were stored in 60-mL (milliliter) serum bottles containing 0.1 milligram of mercuric chloride. Prior to sealing, the serum bottle was flushed with ultra-pure nitrogen before a septum was installed in a manner that minimized entrainment of atmospheric gases into the sample bottle. To collect a sample, an empty auxiliary syringe was inserted through the septum of the serum bottle and 10 cubic centimeters of nitrogen was removed to create a vacuum in the serum bottle. The needle of the collection syringe containing the ground-water methane sample then was inserted through the septum into the serum bottle. Through pressure equilibration, about 10 mL of sample in the sample-collection syringe was pulled into the serum bottle. At two sites, duplicate samples were taken to assess variability. Samples were packed on iced and refrigerated prior to analysis.
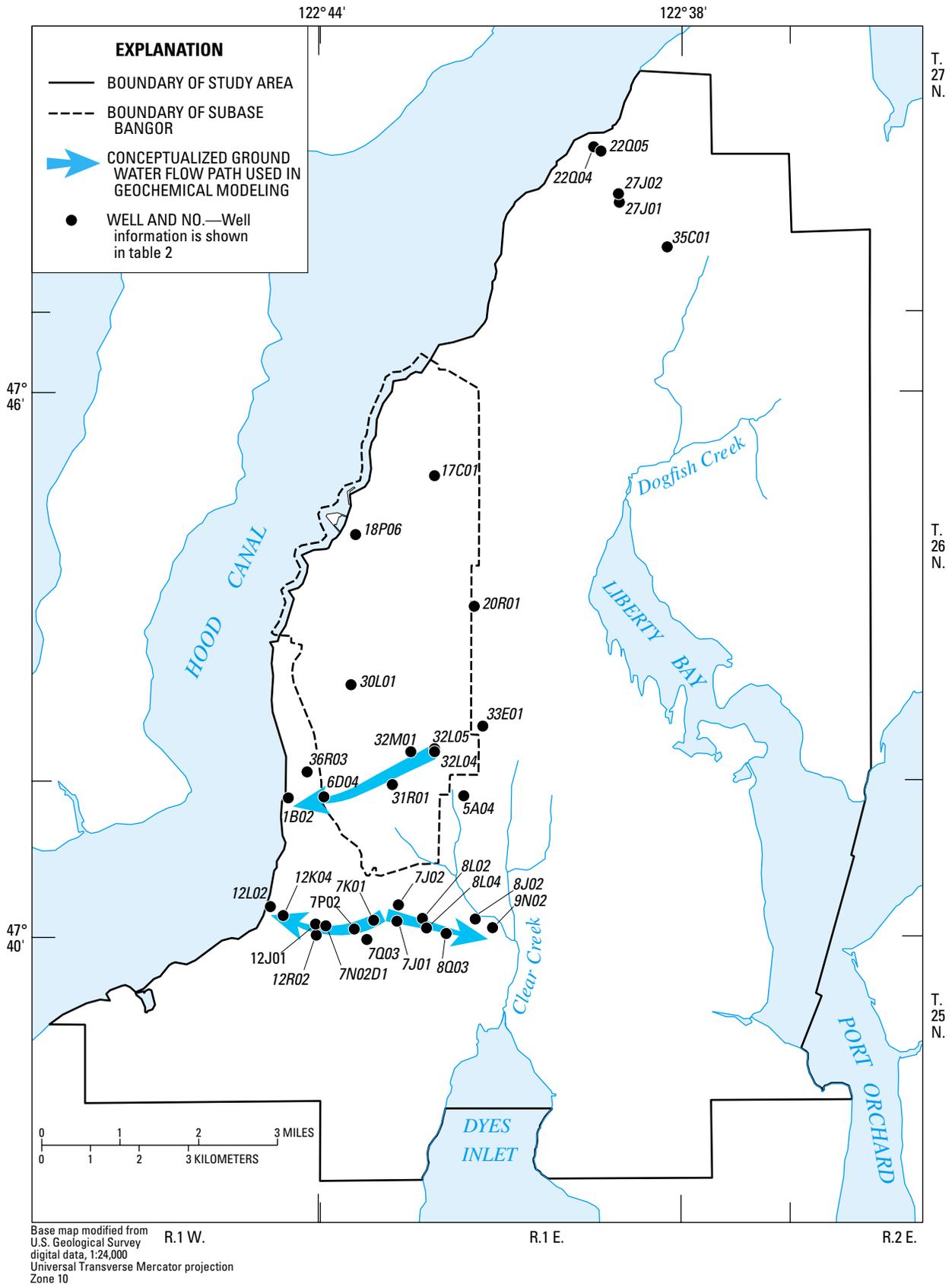
Figure 4. Location of wells sampled for physical properties, dissolved constituent concentrations, and isotopes used to estimate residence time of ground water, and conceptualized ground-water flow paths, SUBASE Bangor and vicinity, Kitsap County, Washington.

Table 2. Location of sampled wells, well-construction data, and hydrogeologic source of water, SUBASE Bangor and vicinity, Kitsap County, Washington

[**Well No.**: Location of wells is shown in figure 4. **Latitude and Longitude**: Degrees, minutes, and seconds. **Altitudes** are given in feet below NGVD29. **Depths** are given in feet below land surface. **Abbreviations:** gal/min, gallons per minute; –, no data]

Well No.	Altitude of static water level (feet)	Discharge rate (gal/min)	Latitude	Longitude	Altitude of land surface (feet)	Depth to top of open interval (feet)	Length of open interval (feet)
Vashon aquifer (Qva)							
25N/01E-05A04	240	13	47 41 31	122 41 43	260	90	8
25N/01E-07J01	186	9	47 40 08	122 42 49	245	73	10
25N/01E-07K01	205	12	47 40 10	122 43 12	320	142	5
25N/01E-08L02	190	12	47 40 10	122 42 24	270	108	5
25N/01E-08L04	207	11	47 40 05	122 42 20	245	65	5
25N/01W-12R02	349	6	47 39 59	122 44 05	425	95	5
26N/01E-20R01	238	11	47 43 35	122 41 32	445	49	5
26N/01E-32M01	254	<.5	47 42 00	122 42 35	322	72	10
26N/01E-33E01	224	30	47 42 18	122 41 24	350	154	5
27N/01E-27J01	172	300	47 48 06	122 39 08	290	161	25
Upper confining unit (QC1)							
26N/01E-17C01	288	0.8	47 45 04	122 42 10	340	131	5
27N/01E-35C01	120	8.5	47 47 34	122 38 21	375	342	5
Permeable interbeds within Upper confining unit (QC1pi)							
25N/01E-07J02	80	8	47 40 19	122 42 47	285	255	5
25N/01E-07N02D1	115	27	47 40 04	122 44 01	425	343	15
25N/01E-07P02	108	15	47 40 03	122 43 31	315	292	5
25N/01E-07Q03	102	7	47 39 55	122 43 18	300	247	5
25N/01E-08J02	87	17	47 40 09	122 41 32	150	163	5
25N/01E-08Q03	90	16	47 40 00	122 42 01	245	260	5
25N/01E-09N02	69	30	47 39 59	122 41 25	60	79	5
25N/01W-12J01	115	11	47 40 07	122 44 03	425	355	5
25N/01W-12K04	53	40	47 40 12	122 44 40	220	199	5
26N/01E-30L01	77	100	47 42 45	122 43 33	325	316	25
27N/01E-22Q04	72	16	47 48 40	122 39 28	120	85	10
Sea-level aquifer (QA1)							
25N/01E-06D04	31	18	47 41 31	122 44 00	294	270	–
25N/01W-01B02	16	9	47 41 30	122 44 35	70	114	5
25N/01W-12L02	2	12	47 40 18	122 44 53	30	81	5
26N/01E-18P06	76	550	47 44 24	122 43 28	175	324	60
26N/01E-32L05	57	300	47 42 02	122 42 11	290	392	0
26N/01W-36R03	11	6	47 41 47	122 44 17	205	276	5
27N/01E-22Q05	22	15	47 48 42	122 39 25	80	234	5
27N/01E-27J02[1]	112	54	47 48 07	122 39 08	290	402	25
Deep aquifer (QA2)							
26N/01E-31R01[2]	27	100	47 41 39	122 42 53	410	512	65
26N/01E-32L04	62	2	47 42 01	122 42 11	285	532	15

[1]Well also may be open to Qva. Analysis of water-quality data indicated that ground water from well 27N/01E-27J02 was likely from the Vashon aquifer. Subsequent efforts to repair well revealed that casing was broken.

[2]Well is screened in QA1 and QA2.

The sample for both carbon isotopes (^{13}C and ^{14}C) was collected in a 1-liter amber borosilicate bottle that had previously been baked at 450°C for at least 2 hours to remove any organic carbon. Prior to collecting the sample, the bottles were flushed for at least 1 minute with nitrogen gas flowing at about 5 liters per minute. The carbon-isotope water sample then was collected by inserting a flexible plastic tube from the filtered water port to the bottom of the bottle and allowing the bottle to overfill by at least three volumes before sealing with a poly-cone cap. Prior to shipping the carbon-isotope samples to the laboratory, 50 mL of sample was removed from each bottle and an equivalent volume of ammoniacal-strontium chloride was added to the sample below the water surface to precipitate the inorganic carbon in solution.

Soil-gas samples were collected for ^{13}C isotope analysis from four heavily forested locations on SUBASE Bangor. The gas samples were collected by driving a 1/4-inch pipe about 4 ft into the soil. A 250-mL gas-sampling bulb was attached to the pipe using flexible plastic tubing. A hand pump was used to pull about 2 liters of soil gas through the gas-sampling bulb before it was closed. The soil-gas samples were sent to the USGS Isotope Laboratory in Reston, Va., for analysis.

CFC samples were collected in triplicate under ultra-pure nitrogen atmosphere and sealed in borosilicate glass ampules by fusing the neck closed using an actylene torch. Because there is a substantial potential for atmospheric contamination when measuring dissolved gases at the parts per quadrillion level (picograms per kilogram), three replicate samples were collected at each location and analyzed individually. The details of the CFC sample collection are described by Busenberg and Plummer (1992).

Sample Analysis

Inorganic ions and nutrients were analyzed at the USGS National Water Quality Laboratory (NWQL) in Arvada, Colo., following the procedures outlined in Fishman and Friedman (1989). Tritium was analyzed at the University of Miami Tritium Laboratory in Miami, Fla., by gas counting following electrolytic enrichment utilizing procedures described by Ostlund and Dorsey (1977). Results of the tritium analysis were reported in picoCuries per liter (pCi/L) and converted to tritium units (TU): 3.2 pCi/L per TU unit. The isotopic ratios of deuterium and ^{18}O were analyzed by mass spectrometry at the USGS Stable Isotope Laboratory in Reston, Va. (Epstein and Mayeda, 1953; Coplen and others, 1991). The isotope ratios were compared to Vienna-Standard Mean Ocean Water normalized to the ratio of Standard Light Antarctic Precipitation (Coplen, 1994). Analytical uncertainty for the isotope measurements (two sigma or two standard deviations) is 0.2 ‰ (per mil, or parts per thousand) for the isotopic ratio of ^{18}O and 2.0 ‰ for the isotopic ratio of deuterium. Methane determinations were performed by head-space gas chromatography using a flame-ionization detector with the associated analytical uncertainty of plus or minus 5 percent.

Tandem accelerator mass-spectrometry analysis was used to analyze the carbon isotope composition of the ground-water samples following preparation of graphite targets. For the five samples collected in 1994, the entire procedure was performed at the University of Arizona Laboratory of Isotope Geochemistry in Tucson, Ariz. For the 20 ^{14}C samples collected in 1995, the graphite target preparations and calculations of the ^{13}C ratios were conducted at the University of Waterloo in Ontario, Canada, and the tandem accelerator mass-spectrometry analyses were performed at the IsoTrace Laboratory in Toronto, Canada. The ^{13}C ratios were determined relative to the Peedee Belemnite (PDB) standard. The ^{14}C results were reported as percent modern carbon (pmc) using ^{14}C standards from the National Institute of Standard Technology (NIST), oxalic acid I and oxalic acid II.

The isotopic ratios of deuterium, ^{18}O, and ^{13}C are expressed in delta notation as δD, $\delta^{18}O$, and $\delta^{13}C$ and are reported in units of per mil. Samples with isotopic ratios greater than the standard have positive delta values and are referred to as "heavy" or "enriched" relative to the standard. Conversely, samples with isotopic ratios less than the standard have negative delta values and are referred to as "light" or "depleted." Additional discussion of isotopic ratios can be found in reports by Clark and Fritz (1997) and Cook and Herczeg (2000).

Analysis of CFCs were performed at the USGS Chlorofluorocarbon Laboratory in Reston, Va., using purge-and-trap gas chromatography with an electron-capture detector as described by Busenberg and Plummer (1992). The results are reported as picograms per kilogram water (pg/kg). The reported detection limit for CFC analysis is 1 pg/kg with an associated uncertainty of 50 percent near the detection level; above 50 pg/kg the analytical uncertainty was plus or minus 3 percent (Ekwurzel and others, 1994). The model recharge year was calculated using the procedures described by Busenberg and Plummer (1992) and a recharge temperature of 9°C. The recharge temperature was selected on the basis of measurements of dissolved nitrogen and argon in three ground-water samples and the seasonal average mean daily temperature for the Bremerton Weather station (near the study area) for the major recharge months of October through May. If CFC concentrations measured in the ground-water samples were greater than maximum reported global atmospheric concentrations of these compounds prior to the time of sample collections, then the concentration data were deemed unsuitable for estimating the residence time of ground water and were labeled as "Concentrations exceed dateable range." The maximum northern hemisphere atmospheric concentrations, measured at Niwot Ridge, Colo., are reported as 294 pg/kg for CFC-11; 570 pg/kg for CFC-12; and 92 pg/kg for CFC-113 (Eurybiades Busenberg, U.S. Geological Survey, written commun., 1998).

As a check for the occurrence of contamination of CFC samples during collection or analysis, water samples were collected from two domestic wells, 25N/01W-01B02 and 12L02 (see fig. 4 for locations), which were expected to be devoid of CFCs. The wells were located near Hood Canal where discharging ground water was expected to be pre-modern. The concentrations of tritium and ^{14}C in samples from these wells were less than 0.3 TU and 80 pmc, respectively, indicating pre-modern ground waters. CFCs were not detectable in the sample from well 25N/01W-01B02, demonstrating that the procedures and equipment used to collect and analyze the CFC samples were non-contaminating. Materials used in well construction and plumbing may have been the source of small concentrations of CFC-11 (2.7-3.0 pg/kg) and

CFC-113 (4.8-6.5 pg/kg) that were detected in the sample from well 25N/1W-12L02. In the plumbing system of well 12L02, a small section of PVC pipe was present ahead of the sampling point, whereas the plumbing system ahead of the sampling point in well 25N/01W-01B02 was constructed entirely of galvanized pipe. Similarly, well 26N/01E-17C01, which was a 2-inch monitoring well constructed entirely of PVC, produced ground-water samples that contained low concentrations of all three CFCs but was classified as pre-modern based on tritium and ^{14}C data. Although the use of PVC and other plastics are demonstrated to be non-contaminating for many organic compounds that are analyzed at the parts per billion concentration level or greater, the presence of plastics and rubber components in well systems to be sampled for dateable concentrations of CFCs (4 to 5 orders of magnitude smaller than parts per billion) is not recommended because of potential sample contamination (L.N. Plummer and Eurybiades Busenberg, U.S. Geological Survey, written commun., notes on procedures for dating young waters, 1994). The wells sampled in this study, particularly wells sampled for CFCs, generally were domestic wells containing pumps and casing components constructed of materials that were not well documented and may have potential for low-level contamination of CFC samples. CFC contamination from well construction or plumbing material was considered a possibility in ground-water samples that contained low concentrations (less than 25 pg/kg) of CFCs or CFC concentrations that generated model recharge years that varied by more than 15 years for the different CFC compounds.

The physical properties and analytical results of chemical analyses were evaluated for internal and external consistency. The common-ion, ORP, and dissolved-oxygen data for well 25N/01E-05A04 were judged to be anomalous with respect to other wells tapping the Vashon aquifer. Information obtained after this well was sampled indicated that the well is located downgradient of the contaminant plume mapped during the remedial investigation of the OU8 contamination site (E.A. Engineering, Science, and Technology, 2000). That investigation concluded that microbial degradation of organic contaminants within the plume was extensive. Because microbial degradation of

organic contaminants can lead to reducing conditions and elevated bicarbonate concentrations like those in samples from well 25N/01E-05A04, it was highly probable that the water samples from well 5A04, although not contaminated, were affected by upgradient land-use activities. All data for this well were included in data tables. However, the dissolved-constituents data were excluded from consideration of the natural variations in the water chemistry of recently recharged ground water in the Vashon aquifer (Qva).

Blanks and replicate samples were submitted as part of the Quality Assurance (QA) program to assess sampling bias and analytical variability. About 15 percent of the samples collected for this investigation were QA samples. With the exception of the CFC data discussed above, results of the QA data indicated the absence of a general bias and low analytical variability, and as such, the data do not need to be qualified for this interpretation. Quality-assurance data for carbon isotope data are tabulated in Appendix A (at back of report) and are discussed where the results affect interpretation of the environmental data.

ESTIMATES OF RESIDENCE TIME AND VARIATIONS IN QUALITY OF GROUND WATER

Estimates of ground-water residence time were determined from the concentrations of environmental tracers measured in samples of ground water collected at selected locations within the ground-water flow system beneath SUBASE Bangor and vicinity. Concentrations of environmental tracers were used to estimate ground-water residence time because the concentration measured in the sample can be related to atmospheric concentrations that were present at the time of ground-water recharge. In the analysis of the differences in tracer concentrations observed between recharge areas and downgradient sampling points, ground-water transport of environmental tracers was characterized as advective flow alone, without consideration of the influences of dispersion or diffusion.

The measured concentrations of environmental tracers in the ground-water samples initially were used to classify the ground waters as either modern (recharged after 1953), pre-modern (recharged prior to 1953), or indeterminate. In ground-water samples that were determined to be pre-modern, and hence older than about 42 years, a radiometric ground-water age based on the concentrations of ^{14}C in dissolved inorganic carbon in the ground water was used to estimate the ground-water residence time. The geochemical model NETPATH was used to refine the ^{14}C estimates of ground-water age by accounting for mass transfers of carbon that occur within the ground-water flow system and that affected the ^{14}C concentration of the sample ground water. Analysis of the spatial distribution of $\delta^{13}C$ ratios, along with concentration data for common ions and other constituents, was used to develop a conceptual model of geochemical processes that alter the chemistry of ground water within the aquifer system present beneath SUBASE Bangor. This conceptual model was used to constrain and select the mineral phases included in the NETPATH model. The stable isotopes of the ground-water samples (oxygen and hydrogen) showed that ground water beneath SUBASE Bangor was recharged under similar climatic and geographic conditions. Physical properties of the water samples (temperature, ORP, and specific conductance) and concentrations of dissolved constituents and isotope ratios were determined for ground-water samples from 33 wells in the study area (table 3). Additional information regarding ambient concentrations of inorganic and organic constituents in the ground water in the study area is available in Greene (1997).

Ground-Water Age Classification Using Environmental Tracers

The tritium, CFC, and ^{14}C data are discussed before the general ground-water chemistry so that general ground-water age relations can be used in the discussion of water-quality variations. In this report, the presence or absence of tritium and CFC is used to distinguish which of the 33 samples contained modern ground water and would not be included for determining ^{14}C model ages for ground water. The chemical characteristic of ground water that contained modern tracers were used to characterize recently recharged ground water and used as the starting solution in developing the geochemical model describing the chemical changes occurring in ground water as it moved through the aquifer system.

Table 3. Physical properties, concentrations of dissolved constituents, and isotope ratios in ground-water samples from wells near SUBASE Bangor, Kitsap County, Washington

[**Well No.**: Location of wells is shown in figure 4. **Bicarbonate**: E, estimated. **Abbreviations**: °C, degrees Celsius; mV, millivolts; μS/cm, microsiemens per centimeter at 25 °C; μg/L, micrograms per liter; TU, tritium units; pmc, percent modern carbon; <, less than; >, greater than; –, no data]

Well No.	Sample date	Temper-ature (°C)	pH (standard units)	Oxidation reduction potential (ORP) (mV)	Specific conductance (μS/cm)	Dissolved oxygen (mg/L)	Calcium (mg/L)	Magnesium (mg/L)
				Vashon aquifer (Qva)				
25N/01E-05A04	07-01-94	10.5	8.3	-180	303	0.2	32	16
25N/01E-07J01	04-19-95	10.1	7.2	>165	105	8.0	8.7	5.7
25N/01E-07K01	03-17-95	8.7	6.9	190	211	5.0	12	16
25N/01E-08L02	03-15-95	9.8	8.3	55	104	8.6	7.6	6.3
25N/01E-08L04	03-11-95	10.1	6.9	150	104	6.2	9.6	4.5
25N/01W-12R02	03-22-95	9.6	6.6	>115	116	8.4	11	5.5
26N/01E-20R01	04-18-95	9.4	7.9	245	110	6.4	6.9	7.2
26N/01E-32M01	04-21-95	10.6	7.2	195	146	5.9	11	9.0
26N/01E-33E01	03-24-95	10.2	7.2	65	183	8.4	13	11
27N/01E-27J01	03-23-95	10.1	7.3	110	184	6.5	14	12
				Upper confining unit (QC1)				
26N/01E-17C01	04-20-95	11.1	8.4	-230	157	<0.1	14	6.4
27N/01E-35C01	03-23-95	9.0	8.6	-150	132	<.1	16	5.8
				Permeable interbeds in Upper confining unit (QC1pi)				
25N/01E-07J02	03-20-95	9.6	7.6	-150	253	<0.1	26	12
25N/01E-07N01D1	03-21-95	9.6	7.8	140	144	3.9	13	7.5
25N/01E-07P02	03-16-95	9.2	7.6	70	158	3.4	13	9.7
25N/01E-07Q03	03-17-95	9.0	7.4	150	162	3.4	13	11
25N/01E-08J02	03-14-95	9.6	7.6	-180	204	.0	23	5.8
25N/01E-08Q03	03-15-95	9.5	8.1	-190	225	.1	23	6.7
25N/01E-09N02	03-20-95	10.9	7.8	-165	209	.0	19	9.1
25N/01W-12J01	03-16-95	9.3	8.1	100	137	9.7	12	6.9
25N/01W-12K04	03-22-95	9.3	7.8	85	121	7.2	12	5.9
26N/01E-30L01	03-24-95	8.7	8.2	-370	160	.2	13	8.8
27N/01E-22Q04	04-18-95	8.5	8.4	-210	184	.4	20	8.1
				Sea-level aquifer (QA1)				
25N/01E-06D04	06-29-94	9.7	7.7	-165	231	0.0	28	7.4
25N/01W-01B02	04-19-95	9.4	7.8	-140	165	.2	19	5.8
25N/01W-12L02	03-20-95	9.0	8.2	125	114	6.5	13	3.9
26N/01E-18P06	03-24-95	9.9	8.0	-140	154	<.1	12	8.6
26N/01E-32L05	03-24-95	9.6	8.2	–	150	.1	16	6.7
26N/01W-36R03	07-01-94	10.6	7.8	-35	154	–	18	6.1
27N/01E-22Q05	04-18-95	9.7	8.3	-95	154	.2	20	3.7
27N/01E-27J02[1]	03-23-95	10.2	7.3	90	218	1.9	18	13
				Deep aquifer (QA2)				
26N/01E-31R01[2]	06-30-94	9.6	7.8	-175	178	.1	20	6.6
26N/01E-32L04	06-28-94	10.4	8.2	-205	149	0.0	15	6.1

[1]Well also may be open to Qva. Analysis of water-quality data indicated that ground water from well 27N/01E-27J02 was likely from the Vashon aquifer. Subsequent efforts to repair well revealed that casing was broken.

[2]Well is screened in QA1 and QA2.

Table 3. Physical properties, concentrations of dissolved constituents, and isotope ratios in ground-water samples from wells near SUBASE Bangor, Kitsap County, Washington — *Continued*

[**Well No.**: Location of wells is shown in figure 4. **Bicarbonate**: E, estimated. **Abbreviations**: °C, degrees Celsius; mV, millivolts; μS/cm, microsiemens per centimeter at 25 °C; μg/L, micrograms per liter; TU, tritium units; pmc, percent modern carbon; <, less than; >, greater than; –, no data]

Well No.	Sodium (mg/L)	Potassium (mg/L)	Chloride (mg/L)	Sulfate (mg/L)	Dissolved inorganic carbon		Silica (mg/L)	Strontium (mg/L)	Barium (μg/L)	Aluminum (μg/L)
					Bicarbonate (mg/L)	Carbonate (mg/L)				
Vashon aquifer (Qva)										
25N/01E-05A04	6.1	2.2	4.2	13	183	0.0	27	140	14	<10
25N/01E-07J01	3.5	0.4	2.2	1.6	60	.0	25	44	3	<10
25N/01E-07K01	5.1	1.0	6.9	2.3	106	.0	39	58	5	<10
25N/01E-08L02	3.6	.5	2.7	0.8	59	.0	28	33	3	<10
25N/01E-08L04	4.2	.4	2.4	2.4	56	.0	24	50	<2	<10
25N/01W-12R02	4.1	.4	3.0	3.4	63	.0	25	49	2	<10
26N/01E-20R01	4.0	.8	2.8	3.5	65	.0	29	37	<2	<10
26N/01E-32M01	4.7	.6	2.0	2.7	84	.0	32	61	3	<10
26N/01E-33E01	6.1	1.4	6.1	3.3	101	.0	30	65	3	<10
27N/01E-27J01	5.6	1.0	2.7	5.8	101	.0	29	80	3	<10
Upper confining unit (QC1)										
26N/01E-17C01	6.9	4.1	1.7	4.7	91	0.0	30	88	12	<10
27N/01E-35C01	7.0	1.9	2.7	3.3	77	.0	32	60	5	50
Permeable interbeds in Upper confining unit (QC1pi)										
25N/01E-07J02	7.1	1.7	1.4	<0.1	166	0.0	47	110	21	<10
25N/01E-07N01D1	4.5	.7	1.8	1.4	89	.0	25	47	<2	10
25N/01E-07P02	4.4	.5	2.4	2.0	93	.0	32	47	4	<10
25N/01E-07Q03	4.5	.7	2.7	1.7	92	.0	30	51	3	<10
25N/01E-08J02	4.7	.8	1.2	1.8	124	.0	42	140	12	<10
25N/01E-08Q03	10	1.6	1.2	1.0	142	.0	39	130	14	<10
25N/01E-09N02	8.1	2.1	1.2	1.1	128	.0	35	89	7	<10
25N/01W-12J01	4.3	1.1	1.8	1.5	71	.0	25	45	<2	<10
25N/01W-12K04	3.8	.5	2.2	2.2	72	.0	23	49	<2	<10
26N/01E-30L01	5.6	1.0	2.1	.4	E103	.0	25	56	5	<10
27N/01E-22Q04	5.5	3.4	2.1	.2	120	.0	36	93	12	<10
Sea-level aquifer (QA1)										
25N/01E-06D04	7.9	1.3	1.4	0.9	149	0.0	37	130	5	<10
25N/01W-01B02	6.7	.5	1.3	.1	109	.0	25	78	6	<10
25N/01W-12L02	4.0	.5	1.7	.9	77	.0	22	56	<2	20
26N/01E-18P06	5.6	1.7	1.9	3.2	89	.0	27	55	4	<10
26N/01E-32L05	5.1	1.5	1.6	2.4	92	.0	30	76	4	<10
26N/01W-36R03	4.7	.4	1.6	1.2	98	.0	22	65	4	<10
27N/01E-22Q05	6.5	1.8	2.0	3.2	92	.0	20	160	5	10
27N/01E-27J02[1]	6.5	1.2	3.6	9.1	115	.0	29	100	3	<10
Deep aquifer (QA2)										
26N/01E-31R01[2]	5.5	1.1	1.3	.4	110	.0	35	90	6	<10
26N/01E-32L04	4.6	1.4	1.5	3.0	93	0.0	27	72	3	<10

[1]Well also may be open to Qva. Analysis of water-quality data indicated that ground water from well 27N/01E-27J02 was likely from the Vashon aquifer. Subsequent efforts to repair well revealed that casing was broken.

[2]Well is screened in QA1 and QA2.

Table 3. Physical properties, concentrations of dissolved constituents, and isotope ratios in ground-water samples from wells near SUBASE Bangor, Kitsap County, Washington — *Continued*

[**Well No.**: Location of wells is shown in figure 4. **Bicarbonate**: E, estimated. **Abbreviations**: °C, degrees Celsius; mV, millivolts; µS/cm, microsiemens per centimeter at 25 °C; µg/L, micrograms per liter; TU, tritium units; pmc, percent modern carbon; <, less than; >, greater than; –, no data]

Well No.	Bromide (µg/L)	Nitrate as nitrogen (mg/L)	Ammonia as nitrogen (mg/L)	Organic nitrogen (mg/L)	Total organic carbon (mg/L)	Dissolved organic carbon (mg/L)	Methane (mg/L)	Phosphate (mg/L)	Boron (µg/L)	Iron (µg/L)
				Vashon aquifer (Qva)						
25N/01E-05A04	0.06	<0.05	0.04	<0.2	0.6	0.3	–	0.03	<10	49
25N/01E-07J01	<.01	.41	<.015	<.2	1.0	.2	–	.02	<10	<3
25N/01E-07K01	.03	2.0	<.015	<.2	–	.4	–	.04	<10	14
25N/01E-08L02	.02	.25	<.015	<.2	–	.3	0.04	.03	<10	12
25N/01E-08L04	<.01	.32	<.015	<.2	–	.3	–	.02	<10	<3
25N/01W-12R02	<.01	.16	<.015	<.2	1.2	.3	–	.02	10	<3
26N/01E-20R01	<.01	.10	<.015	<.2	.3	.2	–	.05	<10	<3
26N/01E-32M01	.02	.75	<.015	<.2	3.5	.3	–	.03	<10	10
26N/01E-33E01	.03	1.0	<.015	<.2	.7	.2	–	.04	<10	<3
27N/01E-27J01	.02	.84	<.015	<.2	.9	.5	–	.04	<10	<3
				Upper confining unit (QC1)						
26N/01E-17C01	0.02	<0.05	0.14	<0.2	1.4	0.4	–	0.12	30	93
27N/01E-35C01	.02	<.05	.04	<.2	.5	.4	0.04	.10	<10	13
				Permeable interbeds in Upper confining unit (QC1pi)						
25N/01E-07J02	<0.01	<0.05	1.6	1.8	–	1.8	23	0.81	30	1,500
25N/01E-07N01D1	.02	.20	<.015	<.2	–	.2	–	.08	<10	<3
25N/01E-07P02	.02	.47	<.015	<.2	–	.3	–	.05	20	22
25N/01E-07Q03	.02	.60	<.015	<.2	–	.6	<.04	.05	20	<3
25N/01E-08J02	<.01	<.05	2.5	2.6	–	1.7	22	1.40	10	800
25N/01E-08Q03	<.01	<.05	1.6	1.7	–	1.8	44	.76	50	650
25N/01E-09N02	<.01	<.05	1.4	1.5	–	1.1	16	.93	30	310
25N/01W-12J01	.02	.25	<.015	<.2	–	.2	–	.08	<10	3
25N/01W-12K04	<.01	.50	<.015	<.2	.4	.2	–	.04	20	<3
26N/01E-30L01	.02	<.05	.16	<.2	2.0	.3	.93	.17	<10	86
27N/01E-22Q04	<.01	<.05	.20	.2	3.2	1.5	–	.22	10	150
				Sea-level aquifer (QA1)						
25N/01E-06D04	<0.01	<0.05	1.3	1.3	1.6	2.0	–	1.50	30	380
25N/01W-01B02	<.01	<.05	.32	.3	2.6	1.8	–	.33	10	650
25N/01W-12L02	.01	.06	.02	<.2	–	<.1	–	.10	<10	<3
26N/01E-18P06	.02	<.05	.14	.3	.6	.2	–	.12	10	55
26N/01E-32L05	.01	<.05	.17	<.2	.4	.4	0.30	.24	10	25
26N/01W-36R03	.01	.10	.02	<.2	.1	.8	–	.10	<10	12
27N/01E-22Q05	<.01	<.05	.14	<.2	.4	.2	–	.12	20	9
27N/01E-27J02[1]	.03	1.2	<.015	<.2	.6	.3	.07	.04	<10	10
				Deep aquifer (QA2)						
26N/01E-31R01[2]	<0.01	<0.05	1.0	1.0	1.2	2.0	–	0.82	10	320
26N/01E-32L04	.01	<.05	.23	.2	.4	.3	–	.30	<10	99

[1]Well also may be open to Qva. Analysis of water-quality data indicated that ground water from well 27N/01E-27J02 was likely from the Vashon aquifer. Subsequent efforts to repair well revealed that casing was broken.

[2]Well is screened in QA1 and QA2.

Table 3. Physical properties, concentrations of dissolved constituents, and isotope ratios in ground-water samples from wells near SUBASE Bangor, Kitsap County, Washington — *Continued*

[**Well No.**: Location of wells is shown in figure 4. **Bicarbonate**: E, estimated. **Abbreviations**: °C, degrees Celsius; mV, millivolts; µS/cm, microsiemens per centimeter at 25 °C; µg/L, micrograms per liter; pmc, percent modern carbon; <, less than; >, greater than; –, no data]
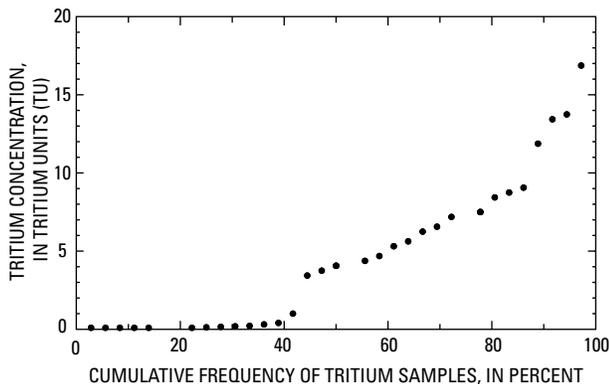
Well No.	Manga-nese (µg/L)	Tritium (tritium units)	Delta carbon-13 (per mil)	Delta deuterium (per mil)	Delta oxygen-18 (per mil)	Carbon-14 (pmc)	Carbon-14 age (years)	Tritium age (years)	CFC age (years)
Vashon aquifer (Qva)									
25N/01E-05A04	100	13	-21.0	-78.9	-11.1	88	<45	<45	–
25N/01E-07J01	<1	4.7	-21.1	-74.9	-10.5	117	<45	<45	Modern
25N/01E-07K01	2	17	–	-77.2	-10.6	–	–	<45	Modern
25N/01E-08L02	1	7.2	-20.3	-77.1	-10.8	115	<45	<45	Modern
25N/01E-08L04	3	3.4	-21.9	-75.1	-10.4	115	<45	<45	Modern
25N/01W-12R02	1	4.1	-21.4	-70.2	-10.4	119	<45	<45	Modern
26N/01E-20R01	<1	14	-18.2	-78.3	-11.1	101	<45	<45	–
26N/01E-32M01	<1	6.2	-24.0	-78.2	-10.8	116	<45	<45	Modern
26N/01E-33E01	<1	8.8	–	-78.2	-10.9	–	–	<45	Modern
27N/01E-27J01	<1	5.3	-21.7	-78.5	-11.0	101	<45	<45	Modern
Upper confining unit (QC1)									
26N/01E-17C01	53	<0.1	-17.40	-79.0	-11.2	60	1,840-2,990	<45	Pre-modern
27N/01E-35C01	33	.2	-18.70	-78.9	-11.2	62	1,900-3,080	<45	–
Permeable interbeds in Upper confining unit (QC1pi)									
25N/01E-07J02	270	<0.1	-12.4	-77.7	-10.8	58	50-510	>45	–
25N/01E-07N01D1	<1	5.6	-21.6	-76.5	-10.9	89	–	<45	–
25N/01E-07P02	<1	7.5	-22.1	-77.7	-10.9	101	<45	<45	Modern
25N/01E-07Q03	<1	9.1	–	-74.3	-10.8	–	–	<45	–
25N/01E-08J02	360	1.0	-10.5	-78.5	-11.0	43	50-1,700	>45	Pre-modern
25N/01E-08Q03	280	<.1	-6.80	-78.7	-11.1	47	920-4,550	>45	–
25N/01E-09N02	91	.2	-15.0	-78.5	-11.0	42	2,130-4,500	>45	–
25N/01W-12J01	<1	8.4	-21.5	-76.4	-11.0	69	–	<45	Modern
25N/01W-12K04	<1	12	-20.4	-78.3	-11.0	86	<45	<45	–
26N/01E-30L01	50	3.8	-21.8	-79.4	-11.2	69	–	<45	–
27N/01E-22Q04	94	<.1	-21.1	-77.7	-11.0	81	–	>45	–
Sea-level aquifer (QA1)									
25N/01E-06D04	440	<0.1	-18.9	-79.2	-11.2	40	330-2,480	>45	–
25N/01W-01B02	72	.3	-13.8	-81.9	-11.4	39	2,750-4,550	>45	Pre-modern
25N/01W-12L02	1	.1	-20.8	-77.4	-11.0	78	–	>45	Pre-modern
26N/01E-18P06	67	7.5	-21.8	-80.3	-11.3	84	–	<45	–
26N/01E-32L05	31	.2	-20.7	-78.1	-11.1	77	50-1,040	>45	–
26N/01W-36R03	<1	<.1	-20.7	-78.8	-11.3	75	50-450	>45	–
27N/01E-22Q05	11	<.1	-18.4	-80.1	-11.2	71	1240-1,480	>45	–
27N/01E-27J02[1]	<1	6.6	-21.2	-76.8	-11.1	96	–	<45	–
Deep aquifer (QA2)									
26N/01E-31R01[2]	180	<0.1	-14.7	-78.9	-11.2	51	180-2,120	>45	–
26N/01E-32L04	28	.4	-20.3	-79.2	-11.2	72	250-1,510	>45	–

[1]Well also may be open to Qva. Analysis of water-quality data indicated that ground water from well 27N/01E-27J02 was likely from the Vashon aquifer. Subsequent efforts to repair well revealed that casing was broken.

[2]Well is screened in QA1 and QA2.

Tritium

Tritium concentrations in ground water from the Bangor area ranged from less than 0.1 to 17 TU (table 3). The laboratory reporting limit for the tritium analysis was 0.1 TU with a standard error of 0.1 to 1.3 TU. Ground-water samples with concentrations less than 0.4 TU are considered to contain little, if any, bomb tritium, indicating that ground water in these samples was recharged prior to 1953. When the tritium data are plotted to show the cumulative frequency distribution (fig. 5), a prominent break in the distribution occurs above concentrations of 1 TU, the level above which the presence of bomb tritium is clearly indicated (Mazor, 1991). Tritium concentrations between 0.4 and 1.0 TU were considered to fall in a range where a clear distinction between modern and pre-modern ground water could not be assigned based on tritium data alone. The reported tritium concentration for wells 25N/01E-08J02 and 26N/01E-32L04 (1.0 and 0.4 TU, respectively) indicate that low concentrations of tritium are likely present in the ground-water samples. Although these data may be interpreted to mean that ground water from these wells recharged in the period just before the start of nuclear weapons testing, a more likely explanation is that the samples are a mixture of predominantly tritium-free pre-modern ground water and a small fraction of modern ground water (on the order of 5 percent) containing bomb tritium. This interpretation is supported by CFC and ^{14}C data, which are discussed later in this section. Similar minor mixing was observed in ground-water samples from the terrace regions of Oklahoma by Busenberg and Plummer (1992).

Chlorofluorocarbons

Concentrations of CFCs were measured in ground-water samples from 14 wells and ranged from less than detection limits to 45,000 pg/kg (table 4). The CFC data indicated that 10 of the 14 wells sampled contained predominantly modern ground water, whereas ground water in the other four wells was predominantly pre-modern. Use of the CFC data to refine the estimates of the ground-water residence time of modern ground water was confounded due to several causes. Concentrations of CFCs exceeded atmospheric equilibrium levels in samples from about 50 percent of the wells sampled. In several of the other wells, there was apparent contamination during sampling or analysis. Lastly, when concentration data were within the dateable range for more than one CFC, widely divergent model ages were produced.

In 7 of 14 wells sampled, the concentration of at least one CFC compound exceeded the concentration attributable to equilibration with the maximum atmospheric concentrations for the northern hemisphere. Model recharge years could not be calculated from these data, which are listed in table 4 as "CEDR, or concentration exceeds dateable range." CFC-12 showed the highest incidence of excessive concentrations, occurring in five wells, where as excessive concentrations of CFC-113 and CFC-11 occurred in four and two wells, respectively. Local sources of CFCs that can create such conditions in ground water include refrigeration equipment, air-conditioners, aerosol spray cans, plastic containers and sewage effluent.

Figure 5. Cumulative frequency distribution of tritium concentration in ground water, SUBASE Bangor and vicinity, Kitsap County, Washington.

Table 4. Concentration of chlorofluorocarbons in ground water and corresponding model of recharge year and ground-water age, SUBASE Bangor and vicinity, Kitsap County, Washington

[**Well No.**: Location of wells are shown in figure 4. **Concentration:** CFC, chlorofluorocarbon. **Model recharge year:** CEDR, concentration of CFC exceeds dateable range; SLLC, suspected low-level contamination. **Concentrations and model recharge years** in parentheses exceed relative-percent-difference limits. **Abbreviations**: pg/kg, picograms per kilogram; >, greater than; <, less than]

Well No.	Concentration (pg/kg)			Model recharge year			Tritium (tritium units)	Estimated ground-water age (years)
	CFC-11	CFC-12	CFC-113	CFC-11	CFC-12	CFC-113		
Vashon aquifer (Qva)								
25N/01E-07J01	171	233	12.2	1969	1982	1974	4.6	Modern
	175	238	(36.3)	1969	1982	(1981)		
	170	239	14.0	1969	1982	1974		
25N/01E-07K01	1.5	37.0	(2.9)	1950	1963	1963	17	Modern
	0.8	35.5	.0	1948	1963	<1955		
	1.2	40.1	.0	1949	1963	<1955		
25N/01E-08L02	31.5	133	5.8	1960	1974	1968	7.1	Modern
	32.8	136	8.0	1960	1974	1970		
	40.2	157	8.3	1961	1975	1970		
25N/01E-08L04	539	41,200	136	1981	CEDR	CEDR	3.4	Modern
	520	41,100	(102)	1980	CEDR	(1991)		
	534	44,900	138	1980	CEDR	CEDR		
25N/01W-12R02	419	470	55	1976	CEDR	1985	4.0	Modern
	421	466	41.5	1976	CEDR	1982		
	(751)	(694)	(80.8)	(1988)	CEDR	1988		
26N/01E-32M01	183	177	5,520	1970	1976	CEDR	6.2	Modern
	191	175	5,990	1970	1976	CEDR		
	195	182	6,180	1970	1977	CEDR		
26N/01E-33E01	392	990	20.5	1975	CEDR	1977	8.6	Modern
	393	1,010	19.2	1975	CEDR	1976		
	(756)	(1,570)	25.0	(1988)	CEDR	1978		
27N/01E-27J01	4,690	3,600	42.3	CEDR	CEDR	1982	5.2	Modern
	4,650	3,550	36.4	CEDR	CEDR	1981		
	9,450	4,420	59.0	CEDR	CEDR	1985		
Upper confining unit (QC1)								
26N/01E-17C01	12.5	1.7	(26.7)	SLLC	SLLC	SLLC	<0.1	Pre-modern
	7.7	8.2	11.9	SLLC	SLLC	SLLC		
	7.0	0.0	16.4	SLLC	SLLC	SLLC		
Permeable interbeds in Upper confining unit (QC1pi)								
25N/01E-07P02	(2,840)	(388)	(90.5)	CEDR	(>1992)	(1989)	7.4	Modern
	105	189	.0	1966	1978	<1955		
	164	256	.0	1969	1983	<1955		
25N/01E-08J02	1.5	.0	6.4	SLLC	<1940	SLLC	1.0	Pre-modern
	.0	.0	.0	<1945	<1940	<1955		
	1.4	.0	.0	SLLC	<1940	<1955		
25N/01W-12J01	2.2	343	4.7	SLLC	1991	SLLC	8.3	Modern
	1.8	343	3.5	SLLC	1991	SLLC		
	1.6	(384)	6.6	SLLC	(1992)	SLLC		
Sea-level aquifer (QA1)								
25N/01W-01B02	0.0	0.0	0.0	<1945	<1940	<1955	<0.3	Premodern
	.0	.0	.0	<1945	<1940	<1955		
	.0	.0	.0	<1945	<1940	<1955		
25N/01W-12L02	3.0	.0	6.5	SLLC	<1940	SLLC	.1	Pre-modern
	2.7	.0	4.8	SLLC	<1940	SLLC		
	(4.7)	(5.0)	(28.8)	SLLC	SLLC	SLLC		

Sampling contamination also may have confounded use of CFC data in several wells. Small concentrations of CFCs, including CFC-113, which was not widely present in the atmosphere prior to 1955, were measured in a sample each, from wells 25N/01W-12L02 and 26N/01E-17C01. Both of these wells contained 0.1 TU or less and less than 80 pmc of ^{14}C (table 3), clearly indicating the samples were pre-modern and that the presence of CFC in the sample was from either contamination or mixing. The presence of CFC in the blank test samples from well 25N/01W-12L02, discussed earlier in the section "Study Methods," demonstrates the presence of contaminants from either well construction materials or procedural problems during sampling or analysis. CFC concentrations less than 25 pg/kg were considered ambiguous with respect to classification of ground-water age unless the calculated model ages for the three CFCs were within 5 years.

Lastly, in the few wells, such as 25N/01E-8L02, which were aerobic and contained concentrations of all three CFCs that were within the range of dateable concentrations, the corresponding model ages were highly variable, further indicating uncertainty in CFC model ages.

Carbon-14

Concentrations of ^{14}C in the 30 ground-water samples from the Bangor area ranged from 39 to 119 pmc (table 3). ^{14}C concentrations in eight wells were greater than 100 pmc, indicating that the sample contained modern water that was recharged during or after the period of thermonuclear weapons testing. Bomb carbon also may be present in ground-water samples with concentrations less than 100 pmc, as indicated by the presence of tritium at concentrations above 1 TU in samples that had ^{14}C concentrations between 80 to 100 pmc.

Concentrations of ^{14}C in samples from the Vashon aquifer ranged from 88 to 119 pmc and concentrations in seven of eight samples were greater than 100 pmc, indicating that ground water in the Vashon aquifer has recharged after the initiation of thermonuclear weapons testing. This conclusion is consistent with tritium and CFC data. The sample in which the ^{14}C concentration was 88 pmc contained 13 TU of tritium, indicating modern ground water. The range of ^{14}C concentrations were similar in samples from the Upper confining unit and the deeper aquifers. Concentrations of ^{14}C ranged from 42 to 101 pmc in the Upper confining unit and permeable interbeds and from 39 to 96 pmc in the Sea-level and Deep aquifers. Smaller ^{14}C concentrations indicate either older ground water, more chemically evolved ground water, or some combination of both.

Concentrations of tritium, CFCs, and ^{14}C were used to classify the age of ground-water samples collected in the study area as modern, pre-modern, or indeterminate (table 5). The primary criterion for classifying a sample as modern ground water was concentrations of tritium greater than 1 TU, concentrations of CFCs greater than 25 pg/kg, and concentrations of ^{14}C greater than 100 pmc. Pre-modern ground water was characterized by non-detectable or trace concentrations of CFCs (less than 25 pg/kg) and tritium (0.4 TU or less) and ^{14}C concentrations less than 85 pmc. When tracer concentration data were within the above ranges, the data were considered unambiguous indicators of the presence of either modern or premodern ground water. Interpretation of ground-water age classification was considered uncertain if concentrations of any one of the environmental tracers was in these ranges: tritium, 0.4-1.0 TU; ^{14}C, 85-100 pmc; CFCs, less than 25 pg/kg. In instances where this occurred, the classification of ground-water age was based on the other tracer data and the ambiguous data were noted in table 5. Indeterminate ground waters were characterized as ground waters containing unambiguous indicators of both modern and pre-modern environmental tracers, such as in well 25N/01W-12J01, which contained 8.5 TU of tritium and 69 pmc of ^{14}C, indicating a mixture of recently recharged and pre-modern ground water.

Table 5. Age classification and concentration of environmental tracers in ground water at SUBASE Bangor and vicinity, Kitsap County, Washington

[**Well No.:** Location of wells are shown in figure 4. **Hydrogeologic unit**: Qva, Vashon aquifer; QC1, Upper confining unit; QC1pi, permeable interbeds within Upper confining unit; QA1, Sea-level aquifer; QA2, Deep aquifer. **CFC recharge**: CFC, chlorofluorocarbon. **Bold** type with V-code indicates near detection level concentrations of some CFCs detected in ground-water samples, which coincided with inconsistent model recharge dates; measured concentrations believed to result from leaching of CFCs from materials used in well construction or plumbing. **Abbreviations:** pmc, percent modern carbon; –, not measured]

Well No.	Hydrogeologic unit	Tritium (tritium units)	Carbon-14 (pmc)	CFC recharge modern/ pre-modern
Modern				
25N/01E-05A04	Qva	13	[1]88	–
25N/01E-07J01	Qva	4.7	117	Modern
25N/01E-07K01	Qva	17	–	Modern
25N/01E-08L02	Qva	7.2	115	Modern
25N/01E-08L04	Qva	3.4	115	Modern
25N/01W-12R02	Qva	4.1	119	Modern
26N/01E-20R01	Qva	14	101	–
26N/01E-32M01	Qva	6.2	116	Modern
26N/01E-33E01	Qva	8.8	–	Modern
27N/01E-27J01	Qva	5.3	101	
27N/01E-27J02[2]	Qva, QA1	6.6	[1]96	–
25N/01E-07P02	QC1pi	7.5	101	Modern
25N/01E-07Q03	QC1pi	9.1	–	–
25N/01E-07N02D01	QC1pi	5.6	[1]89	–
25N/01W-12K04	QC1pi	12	[2]86	–
Pre-Modern				
26N/01E-17C01	QC1	<0.1	60	**Vpre-modern**
27N/01E-35C01	QC1	.2	62	–
25N/01E-07J02	QC1pi	<.1	58	–
25N/01E-08J02	QC1pi	[1]1	43	**Vpre-modern**
25N/01E-08Q03	QC1pi	<.1	47	–
25N/01E-09N02	QC1pi	.2	42	–
27N/01E-22Q04	QC1pi	<.1	81	–
25N/01E-06D04	QA1	<.1	40	–
25N/01W-01B02	QA1	.3	39	Pre-modern
25N/01W-12L02	QA1	.1	78	**Vpre-modern**
27N/01E-22Q05	QA1	<.1	71	–
26N/01E-32L05	QA1	.2	77	–
26N/01W-36R03	QA1	<.1	75	–
25N/01E-31R01[3]	QA1, QA2	<.1	51	–
26N/01E-32L04	QA2	[1].4	72	–
Indeterminate				
25N/01W-12J01	QC1pi	8.4	69	Modern
26N/01E-30L01	QC1pi	3.8	69	–
26N/01E-18P06	QA1	7.5	84	–

[1]Ambiguous indicator concentration, classification not possible without additional information.

[2]Well also may be open to Qva. Analysis of water-quality data indicated that ground water from well 27N/01E-27J02 was likely from the Vashon aquifer. Subsequent efforts to repair well revealed that casing was broken.

[3]Well is screened in QA1 and QA2.

All ground-water samples from the Vashon aquifer (Qva) contained tritium at concentrations ranging from 3.4 to 17 TU, suggesting that ground water in the Vashon aquifer was largely recharged after 1953 (table 5). The presence of CFCs in all samples from Vashon aquifer wells further supports this conclusion (table 4), as do the ^{14}C data, in which seven of eight samples were greater than 100 pmc (table 3). The multiple lines of evidence, tritium, CFC, and ^{14}C, clearly indicate that ground water in the Vashon aquifer recharged after 1953.

Modern ground waters also were present in deeper hydrogeologic units at several locations in the Bangor area. Water in six wells in the QC1pi unit had tritium concentration greater than 1 TU, although only four of those wells (25N/01E-7P02, 7Q03, 7N02D1, and 25N/01W-12K04) were classified as having modern ground waters based on supporting ^{14}C and CFC data. Water in wells 25N/01W-12J01 and 26N/01E-30L01 in the QC1pi unit, which also had concentrations of tritium greater than 1 TU, contained concentrations of ^{14}C at 69 pmc and were thus classified as indeterminate. Water from two deeper wells (27N/01E-27J02 and 26N/01E-18P06), which were both screened in the QA1 aquifer, produced ground water characteristic of modern water, suggesting very rapid recharge deep into the aquifer system. However, for both wells alternate explanations are equally plausible. In well 27N/01E-27J02, which had a reported screen depth of 402 ft below land surface and was classified as a Sea-level aquifer well, the tracer concentrations and overall ground-water chemistry closely resembled well water from the nearby (within 100 ft) shallow well 27N/01E-27J01, suggesting that a preferential pathway was allowing shallow ground water to move rapidly to the deeper well 27N/01E-27J02. Subsequent maintenance work on well 27N/01E-27J02 by KPUD revealed a broken casing that permitted ground water to move between the aquifers both within the well bore and around the outside of the casing (Martin

Sebren, Kitsap County Public Utility District, written commun., 2002). The nearly identical chemistry of water in wells 27N/01E-27J02 and 27J01, which was screened only in the Qva, indicated that ground water from 27N/01E-27J02 should be classified as modern. In contrast, the overall chemistry of water in samples from Sea-level aquifer well 26N-01E-18P06 were characteristic of pre-modern ground water from the deeper aquifer units, but also had substantial concentrations of tritium (7.5 TU) and ^{14}C (84 pmc). This well is a large-capacity production well (500 gal/min) and is part of a well field of six to eight similar wells. The presence of modern water in this well, as evidenced by high tritium concentrations, can be explained by either increased downward flow induced by a substantial increase in the downward hydraulic head gradient produced by the pumping wells or by leaky well construction. Data from this well could not be readily classified as modern or pre-modern and thus was classified as indeterminate.

Samples from seven wells from QC1 and QC1pi had either no detectable or only trace concentrations of tritium and concentrations of ^{14}C less than 85 pmc. The concentrations of CFCs measured in four of these wells were either non-detectable or at trace levels attributable to materials used in well construction or the pump and water discharge lines. These wells were classified as pre-modern ground water (table 5). Pre-modern ground-water samples from the QC1 and QC1pi generally were collected either near discharge areas or at greater depths in the upland areas. Water samples from eight wells tapping the Sea-level aquifer (QA1) and Deep aquifer (QA2) were classified as pre-modern, having no detectable concentrations of tritium and concentrations of ^{14}C less than 85 pmc (table 5). CFCs were not detected in well 25N/01W-01B02, and the trace concentrations in well 25N/01W-12L02 were attributable to materials used in well construction or the pump and water discharge lines.

Variations in Ground-Water Quality

The oxygen and hydrogen isotopic composition of ground water from the SUBASE Bangor area ranged from –11.4 to –10.4 ‰ for δ^{18}O and from –81.9 to –70.2 ‰ for delta deuterium. The isotope data plot as a single group (fig. 6) slightly offset from the global meteoric water line of Rozanski and others (1993). The data generally plot within the range of winter-spring precipitation samples collected at the Global Network of Isotopes in Precipitation site at Victoria, British Columbia (International Atomic Energy Agency, 2001). The Victoria isotope data were collected mostly from 1975 to 1982 at a station located near sea level about 60 miles northwest of the Bangor study area. All of the ground-water samples plot within or near the range of modern precipitation, indicating that they were recharged under climatic conditions very similar to present day conditions and thus implying an upper limit for the age of the ground-water samples as post-glacial, or less than about 10,000 years.

Although the ground-water isotope data plot as a single group, slight differences are apparent between the different hydrogeologic units. Differences in the median concentrations of δ^{18}O from the hydrogeologic units are about 0.2 ‰ and are not large enough to signify distinctly different recharge areas within the irregular geography of the Puget Sound region. The observed variation in δ^{18}O in study wells tapping the Vashon aquifer (Qva) was about 0.7 ‰, which is several times larger than differences between hydrogeologic units. Variations in δ^{18}O due to differences in altitude of precipitation typically are about 0.2 to 0.5 ‰ per 330 ft of change in altitude (Clark and Fritz, 1997). Measurements of δ^{18}O in snow taken from different altitudes on Blue Glacier in the Olympic Mountains indicate a gradient of about 0.5 ‰ per 330 ft elevation (Sharp and others, 1960).

Ground waters characterized as pre-modern appear to have stable isotope ratios of δ^{18}O that are slightly more depleted, about 0.5 ‰, than ground waters characterized as modern, suggesting that these ground waters were recharged under slightly cooler climatic conditions. This difference is not nearly as large as differences observed by Gonfiantini and others (1974) and Hendry and Wassenaar (1999) between ground waters recharged during the present climatic regime and ground waters recharged during the Pleistocene glacial climatic regime. The more-depleted isotope ratios of the pre-modern ground water from the Bangor study area may be the result of recharge during slightly cooler times, such as when the climatic conditions were similar to the Little Ice Age of the late 1800s. An alternative explanation may be that small amounts of Pleistocene ground water containing depleted δ^{18}O have been retained within the fine-grained sediments of the QC1pi and are slowly diffusing into ground water flowing through this unit.
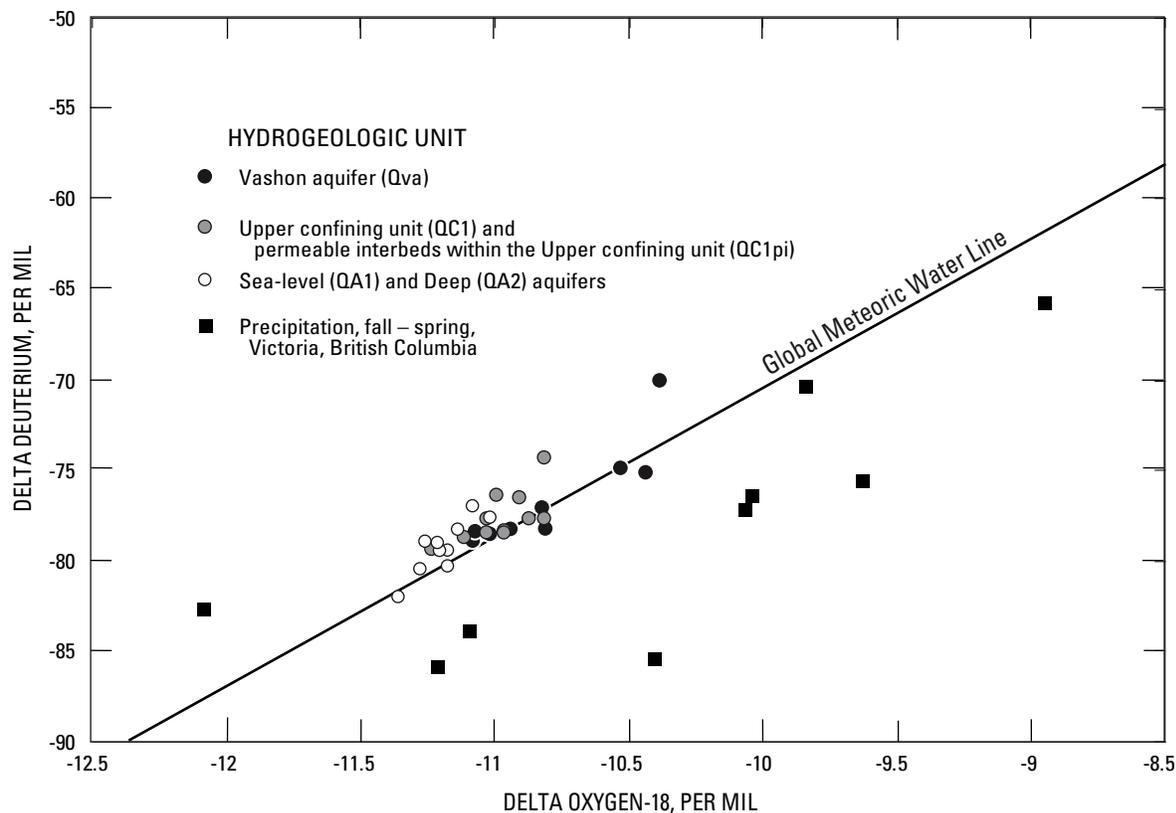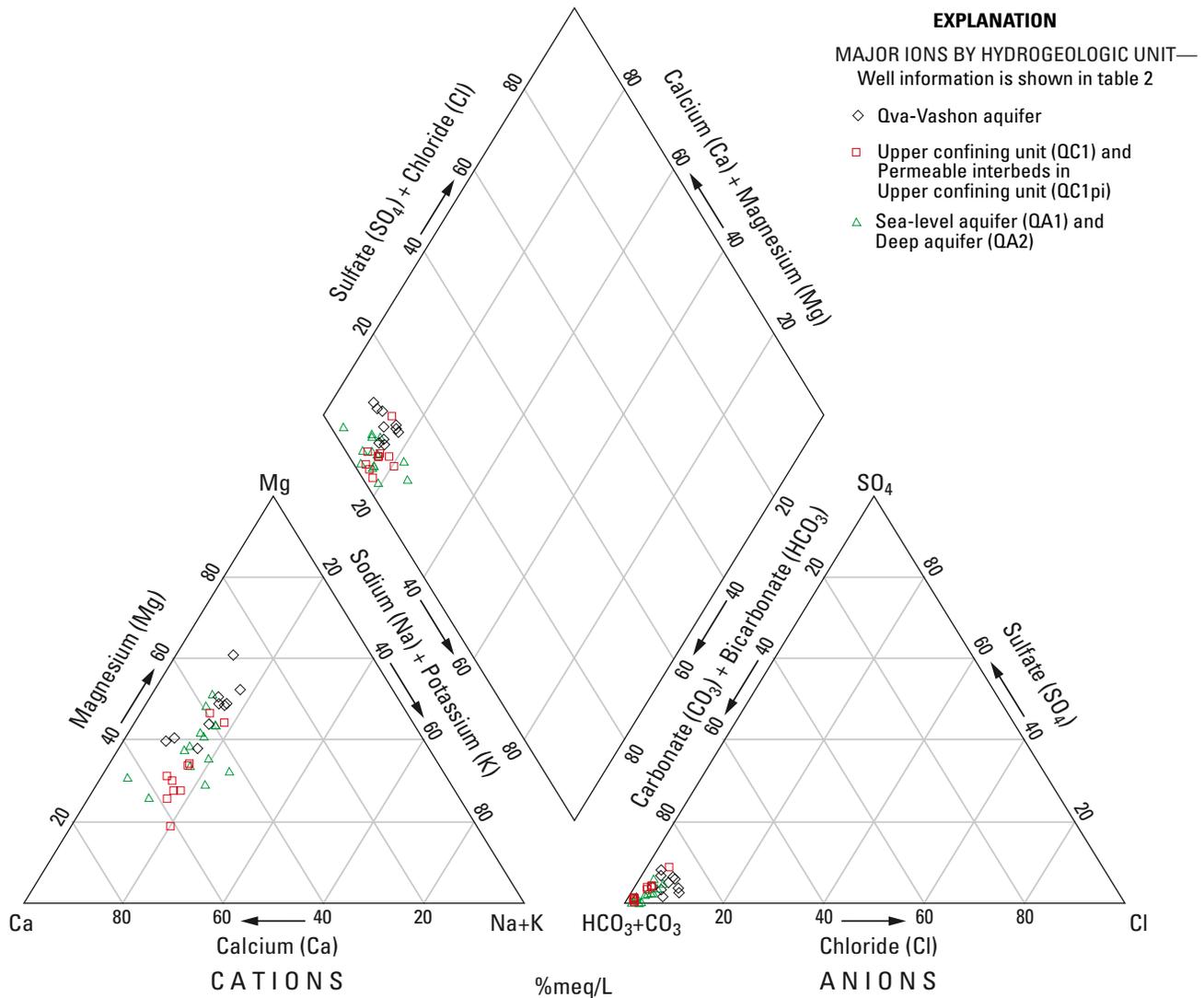
Figure 6. Comparison of stable isotope composition (deuterium and oxygen-18) of ground-water samples and of non-summer (fall-spring) precipitation at Victoria, British Columbia, and the relation with the Modern Global Meteoric Water Line of Rozanski and others (1993), SUBASE Bangor and vicinity, Kitsap County, Washington.

Similar slightly depleted $\delta^{18}O$ ratios have been observed in ground water from aquifers beneath the pre-Vashon confining unit (analgous to the QC1 of this report) in other parts of the Puget Sound region (Stephen Cox, U.S. Geological Survey, unpub. data, 2002).

Ground-Water Chemistry

Concentrations of dissolved constituents in 33 ground-water samples from the Bangor study area were characterized as dilute. In wells unaffected by land-use activities, the specific conductance of ground water ranged from 105 to 253 μS/cm (microsiemens per centimeter). Concentrations of dissolved constituents were smallest in the upland recharge areas, where specific conductance of ground water was about 120 μS/cm, and increased in samples collected from downgradient locations. Based on their major-ion chemistry, these ground waters were classified as predominantly calcium-magnesium/bicarbonate

ground water. Calcium and magnesium together make up more than 80 percent of the cations in solution, with calcium generally present in larger concentrations than magnesium, particularly in wells downgradient from the recharge area. Bicarbonate makes up more than 95 percent of the anions present in ground-water samples. The distribution of major ions in ground water from the Vashon aquifer, the Upper confining unit and permeable interbeds, and the Sea-level and Deep aquifers-were plotted on a trilinear diagram (fig. 7A). The distributions of cations and anions plot in overlapping regions, indicating similarity in the chemical composition of ground water in these hydrogeologic units. The distribution of common ions in these ground waters and their relations with ground-water residence time are consistent with ground waters that have a similar source of recharge and undergo modest chemical evolution of the major-ion composition as ground water moves along a flow path. However, the distribution of cations suggests there may be a subtle difference between ground water from the Vashon aquifer (Qva) and deeper (QA1 and QA2)

EXPLANATION

MAJOR IONS BY HYDROGEOLOGIC UNIT—
Well information is shown in table 2

◇ Qva-Vashon aquifer

□ Upper confining unit (QC1) and
Permeable interbeds in
Upper confining unit (QC1pi)

△ Sea-level aquifer (QA1) and
Deep aquifer (QA2)

A. Distribution of major ions by hydrogeologic units.

Figure 7.   Distribution of major ions in ground water by hydrogeologic units and by ground-water age classification, SUBASE Bangor and vicinity, Kitsap County, Washington.

aquifers. The apparent difference may be an artifact of the limited number of samples from each hydrogeologic unit, or may be due to slight changes in the chemical composition of ground water that typically result from its interaction with the surrounding aquifer material.

Chemical differences in the distribution of major ions were more clearly visible when the major-ion distributions in ground-water samples were grouped on the basis of ground-water age classification (fig. 7B). In most ground waters that were characterized as modern, the magnesium fraction of cations was greater than 40 percent, whereas in pre-modern samples the

magnesium fraction was never greater than 40 percent. Indeterminate ground waters plotted at intermediate positions on the trilinear diagram. The distribution of major ions by age classification shows a pattern of ground-water evolution in which the composition of major ions in older ground waters is increasingly dominated by calcium and bicarbonate. Differences in the chemical composition of ground water are more likely a function of the length of time that ground water is in contact with the aquifer material than of the occurrence of ground water within a specific hydrogeologic unit.
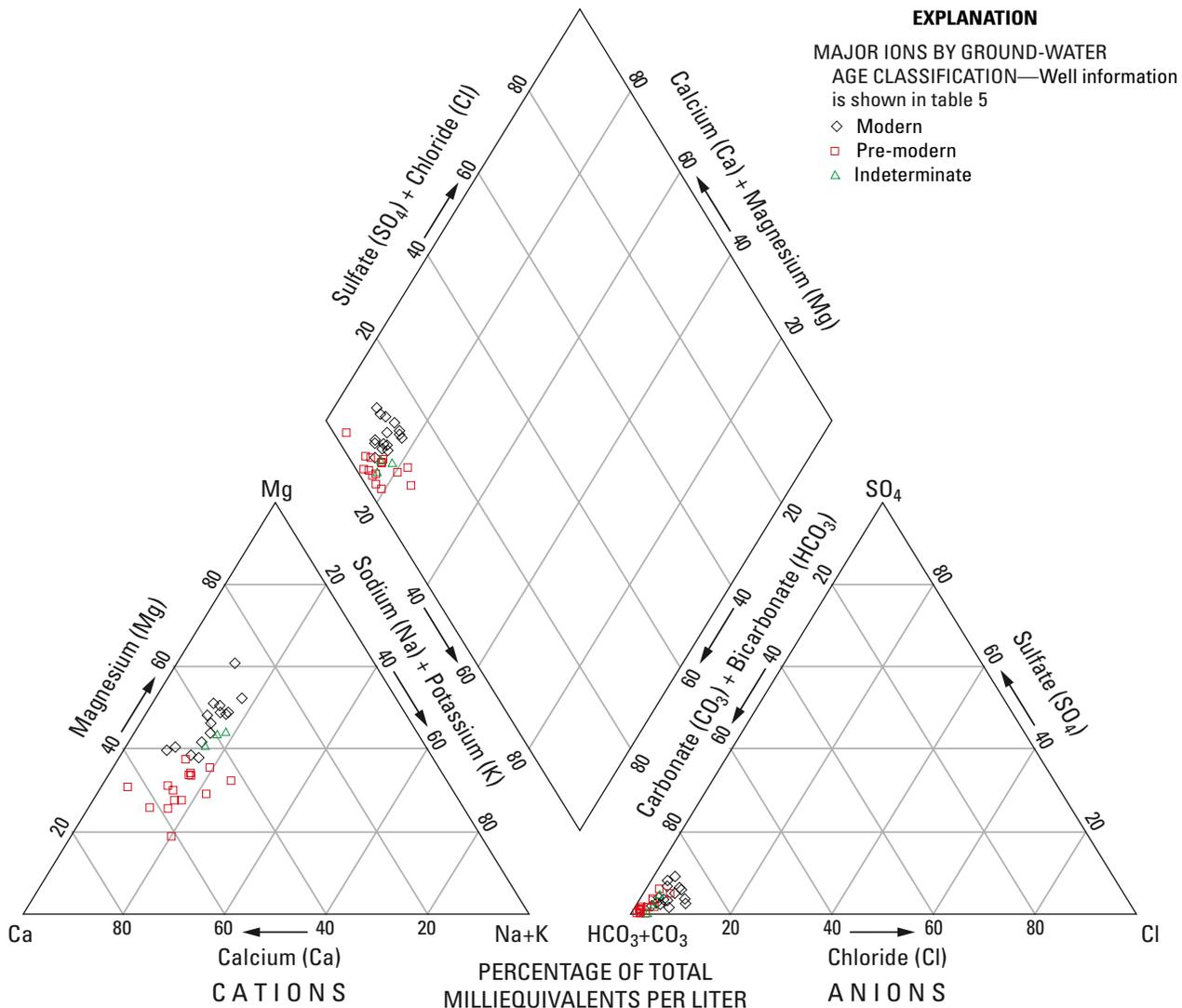
B. Distribution of major ions by ground-water age classification.

Figure 7.—*Continued.*

The common ions that show the largest range in concentration were calcium and bicarbonate. A plot of molar concentrations of calcium and bicarbonate in samples from the 33 wells (fig. 8) shows a linear pattern in which the concentration of bicarbonate is consistently about four times larger than the concentration of calcium. The four to one ratio of the concentration of bicarbonate to calcium is maintained in samples from different hydrogeologic units as well as in the sample from well 25N/01E-5A04 where land-use activities are believed to have affected the overall chemistry of ground water near that area. The

consistency in the ratio of bicarbonate to calcium suggests a similar source material or mechanism for the presence of these constituents. ^{13}C isotope data indicate that the dissolution of calcite, a common calcium-carbonate mineral, is not the source of these ions. Samples with the largest and smallest concentrations of calcium and bicarbonate were both obtained from wells tapping the Qva, although aquifer conditions were considered to be quite different in each case. Concentrations were diluted in samples from wells located in areas where recently recharged ground water could be expected.
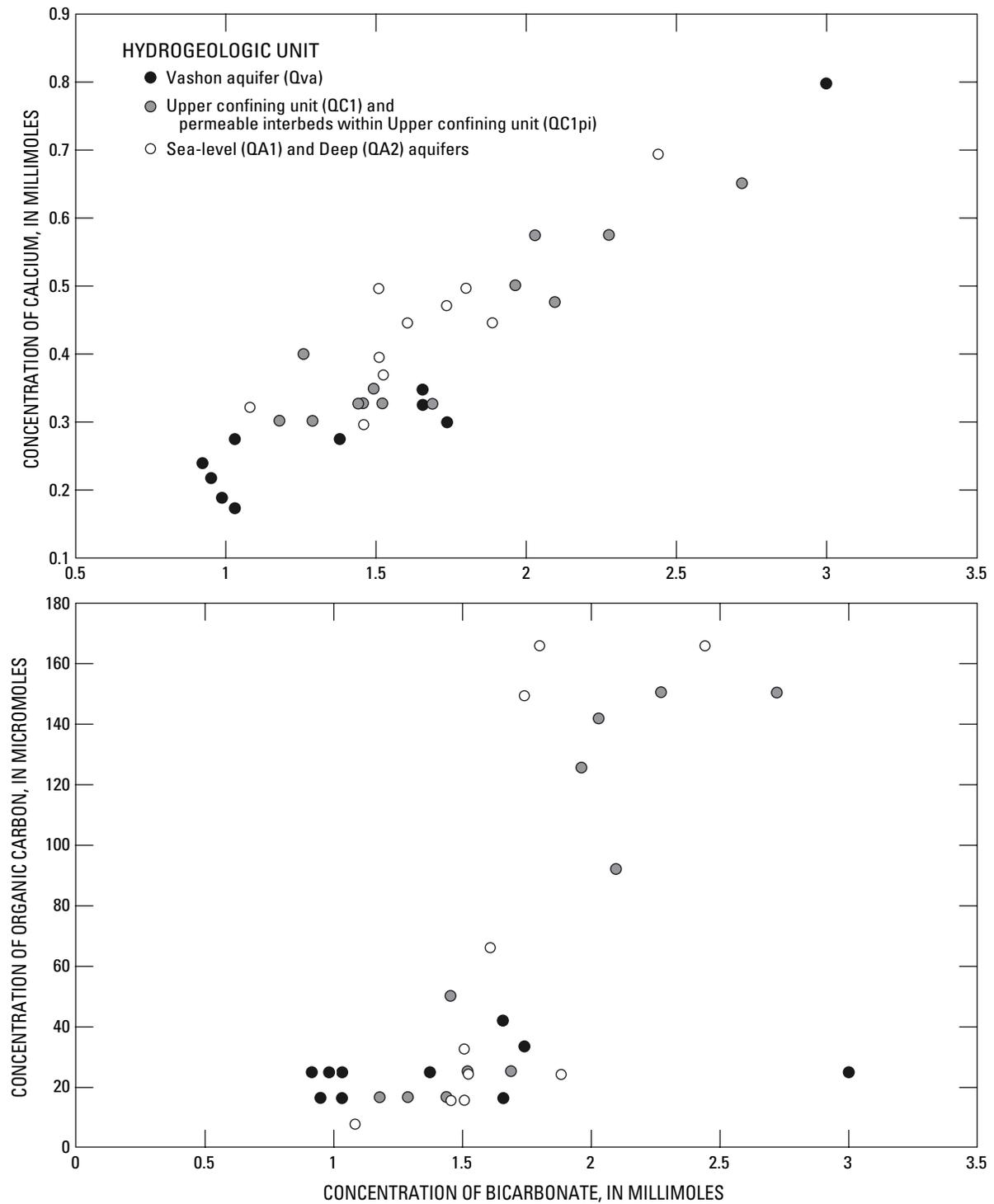
Figure 8. Comparison of molar concentrations of calcium, organic carbon, and bicarbonate by hydrogeologic units in ground water beneath SUBASE Bangor and vicinity, Kitsap County, Washington.

Concentrations were highest in the sample from well 25N/01E-5A04, which was located downgradient of a location where ground-water contamination was found, indicating that land-use activities may have substantially altered the shallow ground-water chemistry. Concentrations of calcium and bicarbonate were often high in samples from wells tapping the QC1pi that were located close to the area of recharge. Concentrations were often smaller in many wells located in what were believed to be downgradient flow-path positions (screened in deeper aquifers or located near ground-water discharge areas), indicating that the ground-water chemistry data cannot be characterized as a generalized progressive change along the inferred ground-water flow paths.

Organic Carbon and Carbon Isotopes

Calculating the model ages of pre-modern ground water based on the radioactive decay of ^{14}C can be straightforward if initial carbon activity is well known and radioactive decay is the only process responsible for decreasing ^{14}C activity. However, ground water from the Bangor area shows a significant increase in carbon as ground water moves through the ground-water system, hence the source and activity of the added carbon must be assessed in order to estimate ^{14}C model ages for these ground waters. Constituents in ground water that are important in understanding the evolution of ground water and the addition of carbon include several different forms of carbon in solution, the concentration of major ions, and the value of the stable isotope ratio of ^{13}C. Carbon species measured in ground water included inorganic carbon, organic carbon, and dissolved methane. Inorganic carbon includes both carbonate and bicarbonate ions in solution (filtered samples) and is referred to as dissolved inorganic carbon (DIC). Only bicarbonate ions were measured in the ground-water samples from the Bangor area. Non-purgeable organic carbon was measured in solution (filtered samples) as dissolved organic carbon (DOC). For about 60 percent of the sites, additional unfiltered samples also were analyzed for total organic carbon (TOC). Dissolved methane was measured in selected samples. The sum of all dissolved carbon species (inorganic, organic, and methane) is referred to as total dissolved carbon (TDC). Isotope ratios of ^{13}C and ^{14}C were measured only on the DIC fraction (bicarbonate) of ground-water samples.

In addition to DIC, other forms of carbon present in ground water beneath SUBASE Bangor included organic carbon and dissolved methane gas. Methane, derived from microbial activity or light hydrocarbon gases, is commonly present in ground water of reduced geochemical aquifer systems (Hem, 1985). Organic carbon, typically present in all natural waters, generally is found at much smaller concentrations than bicarbonate carbon, but can have a substantial effect on the overall ground-water chemistry because of its influence on the solubility of trace elements, potential to participate in redox reactions, and use as a source of carbon and energy to microbially mediated processes that can greatly alter the chemistry of ground water (Hem, 1985). TOC and DOC may act as reactive intermediates in the degradation of sedimentary organic matter to bicarbonate. In samples analyzed for DOC and TOC, concentrations of TOC typically were two to three times larger than concentrations of DOC, indicating that the amount of TOC present in these ground waters often was slightly larger than the amount of DOC but generally of the same order of magnitude. DOC is a better measure of chemically reactive forms of carbon in solution than TOC, which also includes large colloidal carbon molecules, carbon fragments, and microbial detritus that are often recalcitrant (Thurman, 1985).

Concentrations of DOC in the Vashon aquifer (Qva) typically ranged from 0.2 to 0.6 mg/L (fig. 9). Concentrations of DOC in the permeable interbeds (QC1pi) and in the Sea-level aquifer (QA1) were bimodal, with about one-half to two-thirds of the samples having concentrations similar to those in the Vashon aquifer. Most of the remaining samples had DOC concentrations from 1.5 to 2.0 mg/L. A similar bimodal distribution in DOC concentrations was observed in 34 samples from the QC1pi by Greene (1997), although the maximum DOC concentration was slightly larger (3.1 mg/L). Most samples with large DOC concentrations were in the permeable interbeds of the Upper confining unit, which was reported to contain interbedded lenses of peat and disperse organic material in some of the silty deposit (Deeter, 1979). These findings are consistent with other studies in which larger concentrations of dissolved organic carbon were often found in ground water from aquifers containing kerogen and other forms of buried organic matter (Thurman, 1985).

Concentrations of organic nitrogen and ammonia in ground waters of the SUBASE Bangor area also typically were larger in ground water with larger DOC concentrations.

In 6 of 10 ground-water samples analyzed for dissolved methane, the reported concentrations were greater than 0.07 mg/L, the maximum level attributable to atmospheric equilibration, indicating a methane source within the aquifer system. Concentrations of methane in the 10 samples ranged from less than 0.04 to 44 mg/L (table 3), with the largest concentrations observed in samples from the QC1pi. Ground-water samples with large concentrations (greater than 10 mg/L) of methane also had concentrations of DOC greater than 1.0 mg/L. Bacterial methanogenesis is suspected because of the relative abundance of organic carbon reported in some of the hydrogeologic units in which there was no evidence of hydrocarbon deposits such as coal or petroleum. Microbial methanogenesis

can result in significant fractionation of the isotope distribution of carbon constituents dissolved in ground water (Games and Hayes, 1976; Grossman, 1997). Methane was observed in similar-aged glacial sediments in Ontario, Canada, where Aravena and others (1995) found that bacterial methanogenesis had a substantial effect on the estimated age of ground water based on ^{14}C data, as did Barker and others (1979).

Values of δ^{13}C for DIC measured in ground water can provide valuable information on the extent to which microbial and geochemical processes affect ^{14}C concentrations in the ground water. Values of δ^{13}C for DIC measured in ground-water samples from 30 wells near SUBASE Bangor ranged from –24.0 to –6.80 ‰, although concentrations of ^{14}C ranged from 39 to 119 pmc (table 3). Analysis of replicate samples indicated analytical uncertainties of about 0.5 ‰ for δ^{13}C and 1 pmc for ^{14}C.
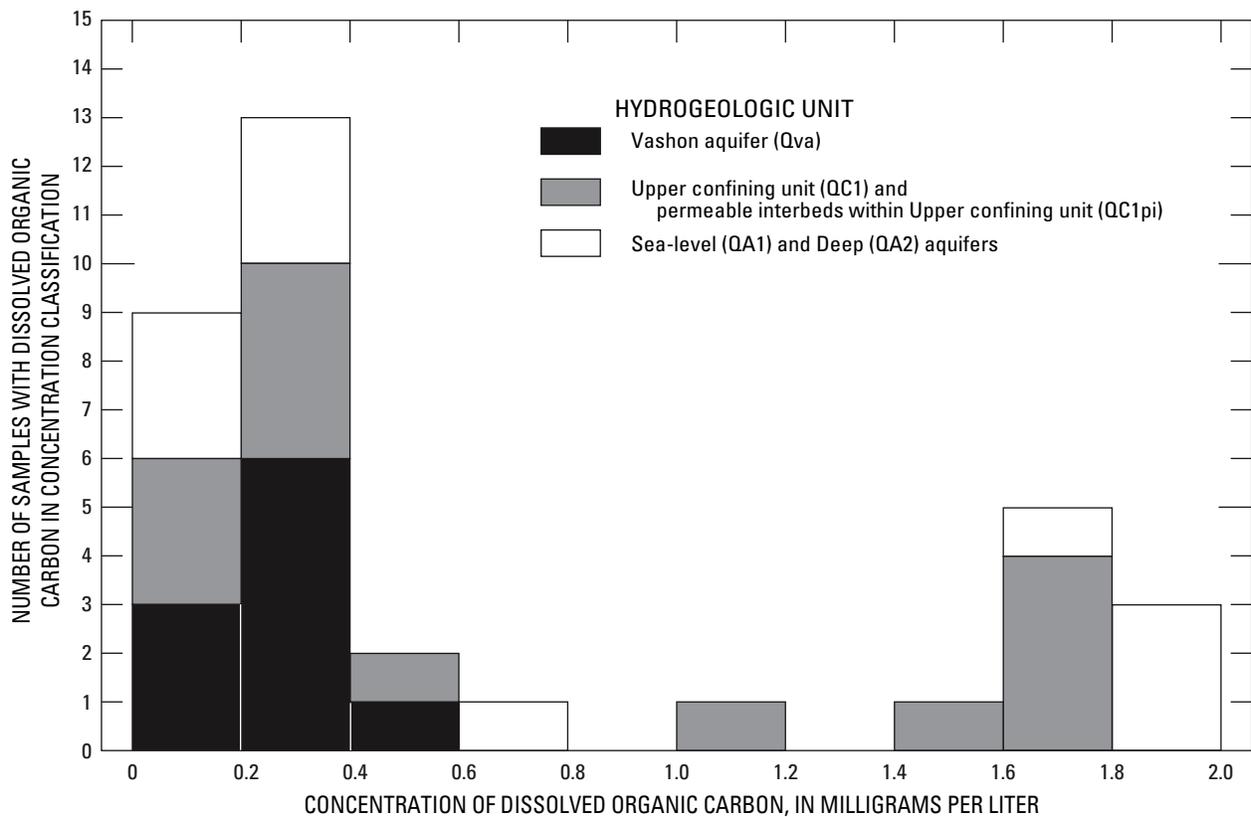
Figure 9. Distribution of dissolved organic carbon in ground water from hydrogeologic units, SUBASE Bangor and vicinity, Kitsap County, Washington.

The $\delta^{13}C$ values of four samples of soil gas collected from heavily forested areas of SUBASE Bangor ranged from –21.6 to –20.6 ‰. The $\delta^{13}C$ ratio of the ground waters characterized as modern ranged from –24 to –18.2 ‰ with a median value of –21.2 ‰, consistent with the $\delta^{13}C$ value in soil gas. The range of $\delta^{13}C$ values present in modern ground waters of the Bangor area is typical of non-carbonate aquifers systems where root respiration and the oxidation of soil organic material is the source of carbon present in ground water (Pearson and Friedman, 1970; Clark and Fritz, 1997). The $\delta^{13}C$ ratio of ground water that was characterized as pre-modern ranged from –21 to –6.8 ‰, indicating that enrichment of $\delta^{13}C$ was occurring in some ground waters downgradient of the Qva.

In 7 of the 30 samples, $\delta^{13}C$ was heavier than –18 ‰ (less negative than –18‰). Methanogenesis and carbonate mineral dissolution are processes that can enrich the $\delta^{13}C$ (cause the value to become less negative) of DIC incorporated into ground water from soil organic-matter sources. Typical $\delta^{13}C$ ratios for carbonate minerals range from –7 to 0 ‰ (Mook, 1980) and the extent of enrichment depends on the amount of carbonate dissolved. Chemical speciation of ground-water samples from the Bangor area indicates that dissolution of calcite was thermodynamically favorable in all ground waters except from well 27N/01E-35C01, and thus if calcite was present in the aquifer sediments, enriched values of $\delta^{13}C$ in DIC (greater than –15 ‰) should have been more prevalent.

During methanogenesis, bacteria fractionate carbon, enriching the $\delta^{13}C$ of DIC and generating highly depleted $\delta^{13}C$ (more negative) in the methane that is formed (Rosenfeld and Silverman, 1959; Barker and Fritz, 1981; and Grossman, 1997). Biogenic methane typically contains $\delta^{13}C$ in the range of –55 to –90 ‰ (Deines, 1980). Aravena and others (1995) evaluated ground-water ages in a methanogenic-glacial aquifer in southern Ontario that also contained carbonate minerals, and found that the DIC in the aged ground waters ranged from –14 to +3 ‰. All ground-water samples from the Bangor study area with $\delta^{13}C$ values greater than –15 ‰ also contained DOC and methane at concentrations greater than 1 mg/L.

The concentration of ^{14}C measured in ground-water samples from 29 wells ranged from 39 to 119 pmc. The occurrence of ^{14}C at concentrations greater than 100 pmc indicates the presence of ^{14}C produced by thermonuclear-weapons testing. In ground-water samples that contained less than 100 pmc of ^{14}C, the concentrations of ^{14}C and ^{13}C vary in a consistent manner such that ground water with diminished ^{14}C concentrations is associated with enriched $\delta^{13}C$ (fig. 10), suggesting that the mechanisms affecting the values of these two isotope concentrations are linked, as in bacterial methanogenesis.

Estimates of the bacterial fractionation of ^{14}C resulting from methanogenesis typically are twice that of ^{13}C (Plummer and others, 1996), thus, the effect of bacterial methanogenesis will be to increase the apparent age of ground water if the age of the carbon being reduced by methanogenesis is older than the carbon present in the ground water. Organic matter present in the Bangor aquifer system has been determined to range from 13,000 to greater than 40,000 years old (Deeter, 1979). Thus, additional DIC in ground water of the Bangor area produced by methanogenic bacteria that are utilizing organic matter containing partially decayed ^{14}C from the aquifer will substantially affect the apparent age of ground water based on the content of ^{14}C in DIC in ground water. Geochemical mass-balance modeling can be used to estimate the amount of additional DIC present in ground water due to microbial methanogenesis.

Changes in Ground-Water Chemistry Along Flow Paths

The composition of dilute ground water typically results from mineral dissolution and chemical alterations brought on by the interaction of the ground water with the aquifer sediments, and results in the accumulation within ground water of dissolved constituents as the water moves along its flow path. If dissolved concentrations rise to exceed mineral saturation levels, then precipitation may occur. Comparisons of the differences in the chemical and physical properties of ground-water samples collected near the recharge and discharge areas of the SUBASE Bangor study area indicate that the chemical evolution of ground water can be characterized by increases in the concentrations of dissolved constituents and the transition from oxidizing to reducing conditions. The largest increases in the concentrations of dissolved constituents were for calcium and bicarbonate, with more modest increases in the concentrations of silica, sodium, potassium, DOC, iron, manganese, and orthophosphate. Oxidizing conditions were common throughout the recharge area, where the concentrations
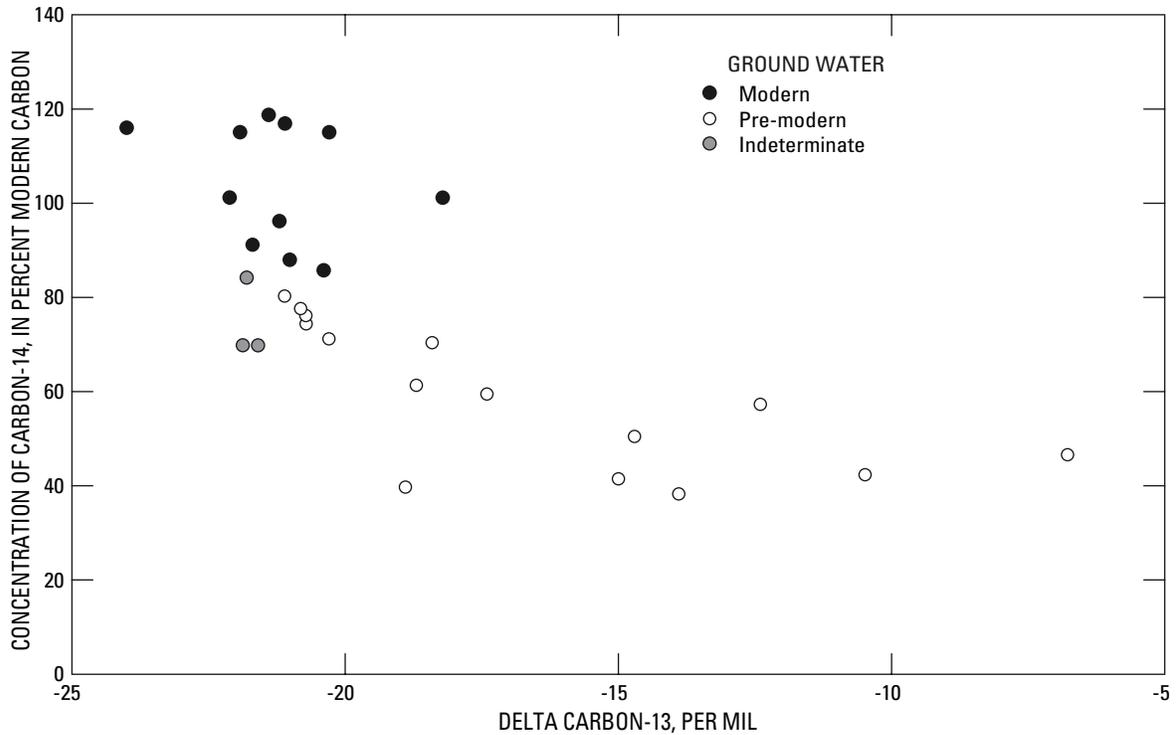
Figure 10. Relation between the value of delta carbon-13 and concentrations of carbon-14 by age classification in dissolved inorganic carbon from ground water near SUBASE Bangor and vicinity, Kitsap County, Washington.

of dissolved oxygen typically were greater than 5 mg/L and the value of ORP was commonly greater than 150 mV (millivolts). Oxidizing conditions rapidly gave way to reducing conditions in the Upper confining unit QC1, as indicated by decreasing concentrations of dissolved oxygen and nitrate and decreasing ORP, typically to less than –150 mV, and increasing concentrations of iron and manganese. At some locations, diminished sulfate concentrations and the presence of methane indicated more extreme reducing conditions. Other changes that were common in some downgradient wells were increases in the concentrations of organic carbon and organic nitrogen and decreases in the concentrations of nitrate and chloride. Concentrations of environmental tracers also were decreasing at sampling sites downgradient of the recharge area. Concentrations of ^{14}C typically were greater than 100 pmc in the recharge area and were generally less than 70 pmc in samples from wells

tapping deeper hydrogeologic units. Likewise, enrichment of δ^{13}C was observed only at downgradient sites.

The most chemically evolved ground waters were those collected from wells tapping the permeable interbeds (QC1pi) or those that moved through the QC1pi to more distant points along the ground-water flow path. The most chemically evolved ground waters are likely related to the presence of organic carbon. Elevated concentrations of calcium and bicarbonate were coincident with elevated concentrations of DOC. For instance, wells 25N/01E-6D04, 7J02, 8J02, and 8Q03, which all had larger concentrations of calcium and bicarbonate than wells at greater distances along the flow path from the recharge area, also had larger concentrations of DOC. Microbial decomposition of organic material within the aquifer may have a substantial effect on the major-ion chemistry in ground water.

Geochemical Mass-Transfer Modeling of Carbon and Estimates of Ground-Water Residence Time

Mass-transfer modeling is a method of accounting for changes in the chemical quality of ground water along a flow path. Measured differences in water chemistry at different locations in the ground-water flow system are accounted for by chemical reactions likely to occur in solution and with minerals of specific elemental compositions thought to be present in the aquifer. Chemical reactions simulated in mass-transfer modeling include dissolution-precipitation of mineral phases, ion-exchange, oxidation-reduction, or gas-exchange. Pairs of samples with different water chemistries were used to represent ground water from different positions in the flow path. The results of this type of modeling typically lead to multiple solutions. Unrealistic solutions can be eliminated by using information on chemical variations in ground water, the thermodynamic solubilities of aqueous species and minerals, and the composition of solid phases within the aquifer. The model also can calculate and predict the isotopic composition of DIC in the ground water resulting from specified mass-transfers, which can be used to simulate the interaction of ground water with the surrounding aquifer material through specific geochemical reactions that fractionate the specific element. This type of modeling provides a method of testing different possibilities for the chemical evolution of ground water, and has been useful in determining the mass transfer of carbon needed to improve estimates of ground-water residence time based on ^{14}C concentrations in the DIC of ground water. Potential sources of carbon in the aquifer of the Bangor area include oxidation of organic matter and the dissolution of carbonate minerals. Potential sources of calcium and silica include weathering of silicate and mafic minerals. Potential sinks for dissolved carbon include precipitation of carbonate minerals and the microbial production of methane.

The modeling procedure used follows the theoretical framework for carbon mass transfer and isotope evolution in natural waters described by Wigley and others (1978; 1979), implemented through the geochemical model NETPATH developed by Plummer and others (1994; 1996). NETPATH accounts for the mass balance of carbon and other constituents by dissolution, precipitation, cation exchange, and oxidation-reduction reactions. Within NETPATH, the

WATEQF subroutine calculates aqueous speciation of elements in solution and the thermodynamic stability of mineral phases (Plummer and others, 1976). Rayleigh distillation equations for fractionating isotopes are used. NETPATH has been used successfully to estimate ground-water residence times in chemically and hydrologically complex aquifer systems where processes such as oxidation of organic matter and outgassing of carbon dioxide are occurring (Aravena and others, 1995; Hinkle, 1995).

Conceptual Model of Ground-Water Evolution near SUBASE Bangor

The selection of mineral phases and chemical constraints used in the NETPATH model was based on a conceptual model of the chemical evolution of ground water in the glacial aquifers and non-glacial confining units in the Puget Sound Lowland. This conceptual model was based on information about variations in the chemical composition of ground water, the composition of aquifer material, and previous geochemical studies in the Puget Sound Lowland, including the work of Dethier (1977), Johnson (1975),Wildrick (1976), Drever and Hurcomb (1986), and Postma and Brockenhuus-Schack (1987). The foundation of this conceptual geochemical model is that frequent precipitation containing dilute concentrations of major ions and charged with dissolved oxygen and carbon dioxide reacts with largely unweathered minerals in sediments deposited during Pleistocene glaciation. The dissolution of soluble minerals such as plagioclase and biotite results in the dilute ground-water solution found in the Qva. The chemical composition of ground water in the Qva develops within the time frame of modern ground water (10 to 50 years) and provides the starting solution for ground waters that continue to evolve chemically in deeper hydrogeologic units. As the flow of ground water proceeds away from the recharge areas, there is an increase in concentrations of carbon (both DIC and DOC), calcium, and magnesium and, to a smaller extent, silica, iron, and ammonia, and a decrease in concentrations of dissolved oxygen, nitrate, and sulfate. In the Upper confining unit (QC1), prominent geochemical processes included the weathering of basalt mineral phases and the oxidation of organic carbon. In some instances, the reduction of carbon dioxide led to the formation of methane via microbial

methanogenesis. Overall, the concentrations of dissolved constituents typically were dilute. Formation of mineral phases by precipitation reactions from solution was limited to secondary clay mineral phases.

Basaltic material in the QC1 is often weathered to the extent that olivine and glass phases have been altered to chlorite and other secondary minerals (Glassley, 1973). General descriptions of basalt weathering indicate that after glass and olivine, the minerals pyroxene and plagioclase are the most susceptible to weathering (Colman, 1982; and Eggleton and others, 1987). Although the weathering of Crescent Formation basalts has not been well characterized, studies of the weathering of Columbia River Basalts demonstrate the alteration of pyroxene to smectite (Hearn and others, 1985; and McKinley, 1990).

The weathering of plagioclase and pyroxenes to kaolinite or smectite is suggested by the molar ratio of sodium to calcium and the ratios of bicarbonate to silica found in the ground water of the Vashon aquifer, which are similar to many other natural waters where these reactions have been studied (Garrels and Mackenzie, 1967; and Drever, 1988). Smectite, montmorillonite, and kaolinite were weathering products identified in glacial sediments of the Puget Sound region and of the Cascade foothills (Wildrick, 1976) and would be consistent with this type of silicate weathering. Evidence of the dissolution of calcite was reported for alpine environments of the Cascade Range by Dethier (1977) and Drever and Hurcomb (1986), but has not been reported in sediments from the Puget Sound Lowland (Mullineaux, 1961; Schlichte, 1968; Johnson, 1975; and Wildrick, 1976).

The increase in both calcium and bicarbonate measured along the inferred ground-water flow path beneath SUBASE Bangor is a common pattern found in ground waters. Often, this pattern is the result of the dissolution of calcite or other calcium-carbonate minerals. This explanation may not be appropriate in the ground-water system of the Puget Lowland, although Drever and Hurcomb (1986) assumed the presence of calcite to explain chemical evolution of waters in glacial sediments of the North Cascade Mountains. Calcite was not observed in the geochemical and mineralogic studies of glacial sediments of the Puget Lowland (Mullineaux, 1961; Wildrick, 1976; and Detheir, 1977) and the $\delta^{13}C$ value of DIC indicates that calcite or other marine carbonates are not a significant source of DIC in ground water of the Bangor aquifer system. Other sources of calcium, such as plagioclase, are widely present in these sediments and they are relatively soluble in dilute ground water, and bicarbonate is a primary product of the oxidation of organic matter.

Organic matter present in the aquifer material can be oxidized and fermented, leading to the formation of bicarbonate. The occurrence of microbial methanogenesis within the aquifer system is indicated by the presence of methane in ground-water samples that characteristically have low ORPs and elevated concentrations of DIC, DOC, and enriched $\delta^{13}C$. The effects of microbial methanogenesis on the value of $\delta^{13}C$ of DIC are large: fractionation factors range from –40 to –75 ‰ depending on metabolic pathway and temperature (Grossman, 1997). The fractionation factor for ^{14}C is estimated to be twice that of $\delta^{13}C$, thus a small amount of methanogenic activity in ground water can have a substantial effect on the apparent ^{14}C age of ground water as well as the value of $\delta^{13}C$.

In this study, ground-water samples with small ^{14}C concentrations have substantially larger amounts of total carbon than those with ^{14}C concentrations near 100 pmc. The major source of additional carbon in the SUBASE Bangor ground-water system is believed to be the oxidation or fermentation of sedimentary organic material. Deeter's (1979) examination of the glacial deposits of the Kitsap Peninsula showed that organic material commonly is found in the fine-grained interglacial deposits. Organic material also has been noted in many well drillers' reports from the area. The increase in concentrations of DOC, nitrogen, and ammonia with decreasing ^{14}C concentrations is consistent with the oxidation of organic material. Microbial oxidation of organic material has been shown to occur deep within aquifers and to result in the increase of bicarbonate concentrations along ground-water flow paths (Chapelle and Knobel, 1985; Chapelle and others, 1987; McMahon and Chapelle, 1991; and Murphy and others, 1992).

NETPATH Modeling of Ground-Water Residence Time

To provide more reliable estimates of the residence time of pre-modern ground waters based on the concentration of ^{14}C in DIC, NETPATH simulations were used to model the geochemical interactions that affect the concentration and isotopic composition of DIC in ground water as it flows between the point of recharge and the point at which the sample was collected. The NETPATH model calculates the ^{14}C concentration at the downgradient well by modifying the ^{14}C concentration found in the upgradient well for chemical reactions occurring along the flow path, and not for radioactive decay. The calculated activity is referred to as the "non-decay ^{14}C concentration." The ratio of the ^{14}C concentration measured in the sample to the non-decay ^{14}C concentration is utilized in the radioactive decay equation to calculate ground-water residence time. Multiple NETPATH simulations were run to evaluate the sensitivity of the modeling results to uncertainty in ^{14}C concentrations of the initial solution and the organic material found in the aquifer sediments.

Mass-transfer modeling using NETPATH was applied to water chemistry reported for 14 downgradient wells in which environmental tracer data indicated that the ground water could be characterized as pre-modern (table 5). A mass-transfer model was not developed for well 25N/01W-12L02 because the DIC concentration in this sample was small, the water contained dissolved oxygen, and the general chemistry resembled recently recharged ground water. To the degree possible, the location of these wells was concentrated in localized areas to provide a cross-sectional view of the distribution of ground-water residence times. The NETPATH simulations were carried out on paired sets of water chemistries from samples representing an upgradient and a downgradient location in the ground-water system. The upgradient conditions for all NETPATH simulations were represented by the median concentrations of ground-water constituents found in wells tapping modern water in the Vashon aquifer (table 5; well 25N/01E-5A04, which was judged to be anomalous, was not used). The median cation and anion concentrations for these nine wells were within a few of percent of the median concentrations for the 54 wells from the Vashon aquifer sampled as part of the ambient water-quality part of the broader study at SUBASE Bangor (Greene, 1997). The charge balance

for the chemical analysis (difference between the sum of median values for anions and cations) was 2.34 percent for the 9 wells and 1.75 percent for the 54 wells, which was acceptable. For the aqueous speciation modeling, the charge balance was reduced to zero by adjusting the concentration of bicarbonate.

A lack of extensive data on the mineralogical composition of glacial sediments limited the extent to which a conceptual geochemical model of the chemical evolution within those sediments could be developed. A specific concern for the correction of ^{14}C estimates of ground-water residence time is the apparent lack of carbonate minerals in glacial sediments in the Puget Sound Lowland. Of the mineralogical and chemical analysis reported by previous investigators, only one reported the presence of siderite, an iron-carbonate mineral, as a minor component in one sample.

Constituents used to constrain the NETPATH model for chemical evolution of downgradient wells included the concentrations of carbon, calcium, sodium, iron, sulfur, and silica and redox state. The following phases were available to the NETPATH modeling process, but only seven phases (equal to the number of constraints) could be included in any specific version of the models developed. Mineral phases included pyroxene (augite), chlorite, pyrite, smectite, mafic montmorillinite, calcite, goethite, and gypsum. Non-mineral phases include organic matter as CH_2O and carbon dioxide and methane gases. Anion exchange of two sodium ions for one calcium ion could also be used as a model phase. The mineral phase gypsum, which is not known to occur in these aquifer sediments, was included to provide a source of sulfate ions required in models for three wells. The source of the sulfate observed in ground water from the deeper hydrogeologic units is not known. Possible explanations include pyrite oxidation, as suggested by Tesoriero and others (2000), or the fine-grained deposits of the adjacent confining unit, as described by Chapelle and McMahon (1991).

With the exception of smectite and chlorite, the stoichiometry of the mineral phases was default values contained in the NETPATH code. Smectite, a mafic montmorillinite, contains a larger fraction of calcium than the default mafic montmorillinite. The stoichiometry of smectite used here was derived from weathered basalts of the Columbia River Basalt Group (Hearn and others, 1985). The analysis of chlorite by Glassley (1973) indicated that iron made up about 40 percent of the cations in mineral formula of chlorite in basalts of the Crescent Formation. Chlorite was

assigned a stoichiometry of $(Mg_{3.5},Fe_{1.5})$ $Al_2Si_3O_{10}(OH)_8$. The NETPATH mass-balance model computed the amounts of each phase required to either dissolve, precipitate, or exchange in order to match the chemical changes observed between the modern ground water, as represented by the median concentrations from wells tapping the Qva, and the pre-modern water, from the 14 downgradient wells. The thermodynamic validity of the dissolution-precipitation reactions was checked within the WATEQF subroutine by examining the saturation index calculated for mineral phases at the downgradient well.

Other carbon isotopic data needed in the mass-transfer modeling were either measured during this study or taken from published values. Carbonate minerals were considered an active phase in three of the models, in which case the isotope values for calcite were $\delta^{13}C = 0$ ‰ and ^{14}C activity = 0 pmc (Clark and Fritz, 1997). With the apparent absence of carbonate minerals in the soils and unsaturated zone, the $\delta^{13}C$ value in the recharge ground water is assumed to be that found in soil gas. The $\delta^{13}C$ of four samples of soil gas ranged from –21.6 to –20.6 ‰. A value of –21.0 ‰ was used in all carbon mass-transfer models. The $\delta^{13}C$ of organic matter in the aquifer was assumed to be –25.0 ‰, which is a commonly used value for plant matter (Mook, 1980). Methane also was generated in 9 of 14 of the mass-transfer models. The $\delta^{13}C$ of methane from ground water of the study area was not measured and was assigned a value of –70 ‰, based on ^{13}C analysis of biogenic methane found in glacial-drift deposits (Deines, 1980; Arevena and others, 1995). The resulting fractionation factor for $\delta^{13}C$ from the methanogenic conversion of $\delta^{13}C$ from –25 to –70 ‰ was calculated to be 1.0541.

The modeling technique utilized by NETPATH will find all possible models utilizing the phases that satisfy the model constraints, and as such NETPATH can generate numerous hypothetical models. Models that violate geochemical constraints, such as precipitation of a phase in which the aqueous concentrations were not saturated, were eliminated from consideration. The primary check on the suitability of models involving the mass transfer of carbon was a comparison of model-calculated values of $\delta^{13}C$ to the values of $\delta^{13}C$ measured in the downgradient wells.

In general, the overall reactions that were modeled by NETPATH to represent the geochemical evolution of ground water between the recharge areas and the sampled well were the oxidation of organic matter, the dissolution of pyroxene (augite) and chlorite, and the precipitation of smectite, montmorillinite, goethite, pyrite, and the exchange of calcium for sodium ions. In 60 percent of the wells, the production of methane was modeled. The modeled mass transfers that occurred between the recharge area and well 25N/01W-01B02 could be written in the following manner:

$$MedQva_{(dissolved)} + 0.82\ CH_2O + 0.82\ pyroxene$$
$$+ 0.06Na_2X \Rightarrow 0.28\ methane + 0.43\ smectite$$
$$+ 0.06\ goethite + 0.02\ pyrite + 0.06\ CaX$$
$$+ Well\ 1B02_{(dissolved)}, \qquad\qquad (2)$$
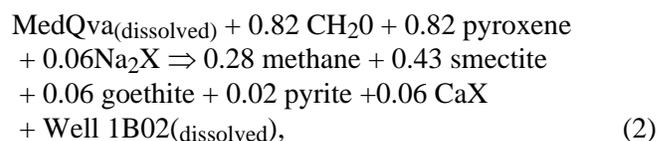
where:

$MedQva_{(dissolved)}$ = median concentration of dissolved constituent in the Vashon aquifer,

$Well\ 1B02_{(dissolved)}$ = concentration of dissolved constituents in well 25N/01W-01B02,

Na_2X = sodium exchange site on solid phases, and

CaX = calcium exchange site on solid phases.

$MedQva_{(dissolved)}$ and $Well\ 1B02_{(dissolved)}$ represent the dissolved constituents present in the initial and final waters, respectively. The reaction coefficients are in millimoles (mmoles) per kilogram of water. For mass balance of carbon between ground water from the Vashon aquifer and well 25N/01W-01B02, 1.41 mmoles of carbon are present in the initial ground water of MedQva, to which is added 0.82 mmole of carbon from CH_2O through the oxidation of organic material. With the production of 0.28 mmole of methane, 1.95 mmoles of inorganic carbon remains in the ground water sampled from well 25N/01W-01B02. Uncertainty relating to the silicate mineral phases led to several modeled solutions. However, the estimates of ground-water residence time were not sensitive to the silicate phases utilized in the model and estimates of ground-water residence time varied by only a few percent under the various silicate mineral scenarios.

Selected results of the NETPATH modeling that represent the geochemical evolution between the recharge area and 14 downgradient sampling locations are shown in [table 6]. The value of $\delta^{13}C$ of ground water DIC was not used as a model constraint in the construction of NETPATH models and was allowed to change on the basis of the geochemical reaction involving carbon. The primary criterion for the selection of models representing mass transfers of carbon and the concurrent effect to ^{14}C was the accuracy in the model-predicted $\delta^{13}C$ compared to measured $\delta^{13}C$. The model that included the oxidation of organic carbon and methanogenesis was found to satisfy the basic mineral and water-quality constraints for all ground waters modeled; however, methanogenesis was invoked to model mass transfers at only nine sites. The values of $\delta^{13}C$ predicted by 9 of the 14 mass-transfer models were within 1 ‰, which is considered a good match. Analytical variability in the measurement of $\delta^{13}C$ is about 0.5 ‰ and measurement of $\delta^{13}C$ in three sets of duplicate ground-water samples varied from 0.3 to 0.6 ‰ (Appendix A). The maximum difference in the model-predicted value of $\delta^{13}C$ and the measured value was 2.5 ‰.

The calculated value of $\delta^{13}C$ of DIC was very sensitive to the amount of methane generated. The largest difference in the modeled and measured values of $\delta^{13}C$ of DIC for methanogenic models was 2.3 ‰ for well 25N/01E-8Q03. Some of the discrepancy may be related to uncertainty in the methane analysis. The quality-assurance data for the methane analysis were limited to replicate samples, which showed little variation but could not be used to assess bias in the analysis. For the five wells with large measured methane concentrations, the model-calculated quantity of methane was typically about one-half or less than the measured amount. Whether this indicates an additional source of methane, inaccurate analysis, or inappropriate modeling could not be determined. However, when the model for well 25N/01E-8Q03 was run without the methane present in the downgradient solution, the difference between the modeled and measured values of $\delta^{13}C$ of DIC was 1.3 ‰.

Variations and Distribution of Carbon-14 Estimate of Ground-Water Residence Time

After the NETPATH models were developed that fit geochemical constraints, ground-water residence time based on ^{14}C concentrations were estimated. A series of model runs were performed to evaluate the effects of uncertainty in two important model input parameters that could not be well constrained: the initial concentration of ^{14}C in the ground water of the recharge area and the concentration of ^{14}C in the organic material present in the QC1 that was the source of additional carbon in ground water.

The range of concentration of ^{14}C measured in modern ground water from the recharge area was large, from 86 to 119 pmc and undoubtedly affected by the ^{14}C generated by the testing of thermonuclear weapons and thus not representative of ^{14}C concentrations present at the time pre-modern ground waters were recharged. The presumption that ^{14}C activity of the recharge was 100 percent modern carbon may not be realistic as the oxidation of old carbon within soils can generate soil CO_2 in which the ^{14}C activity is less than modern (Clark and Fritz, 1997). Likewise, pediogenic carbonates and kerogen incorporated in glacial till (Vogel, 1967; Keller, 1991) also could affect the concentration of ^{14}C without affecting the value of $\delta^{13}C$. Coal and shale from bedrock overridden by moving glaciers are the reported sources of the kerogen found in till by Keller (1991). These sources of carbon are not extensive in the Puget Sound region, although they are found in some areas. Vogel (1967) assigned an initial ^{14}C concentration in a glacial aquifer of 85 pmc to account for additional DIC incorporated into recharging ground waters from carbonate minerals in the unsaturated zone. Preliminary estimates of ground-water residence times using data from downgradient wells of this study and Vogel's initial ^{14}C concentration of 85 pmc resulted in negative estimates of ground water residence time in some wells. Thus, using 85 pmc for the initial activity produced overestimations of the amount of DIC dilution in the initial ground waters at some wells but not others. Kalin (2000) reported that oxidation of Holocene organic matter resulted in ^{14}C activity from 88 to 92 pmc in recently recharged ground water which also contained tritium.

Table 6. Mass transfers of modeled phases and measured and model-calculated values of carbon-13 from selected geochemical mass-transfer models of pre-modern ground water, SUBASE Bangor and vicinity, Kitsap County, Washington

[**Well No.:** Location of wells is shown in figure 4. **Phases**: Mass transfer units are in millimoles per liter. Values in parentheses indicate concentration in solution. **Hydrogeologic units**: QC1, Upper confining unit; QC1pi: permeable interbeds within Upper confining unit; QA1, Sea-level aquifer; QA2, Deep aquifer. **Abbreviations**: –, phase not part of model]

Methanogenic models

Phase	Hydrogeologic unit and well No.								
	QC1	QC1pi				QA1			QA1, QA2
	26N/01E-17C01	25N/01E-07J02	25N/01E-08J02	25N/01E-08Q03	25N/01E-09N02	25N/01E-06D04	25N/01W-01B02	27N/01E-22Q05	26N/01E-31R01[1]
Ca/Na exchange	0.062	0.084	0.019	0.226	0.108	0.074	0.063	0.061	0.035
Goethite	-.767	-2.33	-.168	-5.06	-.993		-.059	-.072	-.122
CH_2O		2.45	1.52	3.76	1.83	1.49	.816	.175	.935
CH_4 gas	-.036	(-1.44)	-.663	(-2.75)	(-1.00)	-.213	-.282	-.067	-.333
Pyrite	–	-.022	-.012	-.017	-.016	-.21	-.021	.005	-.02
Calcite	–	.578	–	–	–	–	–	–	–
Chlorite	.362	1.09	.88	2.24	.476				
Augite	–	–	.958	2.45	1.09	.327	.819	.901	.789
Smectite	-.284	-.798	-.429	-3.03	-.851	–	-.434	-.499	-.373
Mafic montmorillinite	–	–	–	–	–	-1.29	–	–	–
SO_4, gypsum	.005	–	–	–	–	.384	–	–	–
Comparison of carbon-13 measured in samples and calculated from mass-balance modeling									
$\delta^{13}C$, measured, per mil	-17.4	-12.4	-10.5	-6.8	-15	-18.7	-13.8	-18.4	-14.7
$\delta^{13}C$, model calculated, per mil	-17.9	-10.4	-10.1	-4.5	-15.4	-18.7	-15.2	-19.3	-14.4

Non-methanogenic models

Phase	Hydrogeologic unit and well No.				
	QC1	QC1pi	QA1		QA2
	27N/01E-35C1	27N/01E-22Q4	26N/01E-32L5	26N/01W-36R3	26N/01E-32L4
Ca/Na exchange	0.068	0.027	0.022	0.009	0.009
Goethite	-.119	-.401	-.129	.02	-.092
CH_2O	.082	.649	.129	.197	.135
CH_4 gas	–	–	–	–	–
Pyrite	-.005	-.147	-.009	-.016	-.006
Calcite	-.127	–		.123	
Chlorite	.	.243	.032		.047
Augite	.799	–	.462	.199	.351
Smectite	-.392	-.161	-.255	-.129	-.208
SO_4, gypsum	–	.252		–	
Comparison of ^{13}C measured in samples and calculated from mass-balance modeling					
$\delta^{13}C$, measured, per mil	-18.7	-21.1	-20.7	-20.7	-20.3
$\delta^{13}C$, model calculated, per mil	-21.2	-22.1	-21.3	-19.8	-21.4

[1]Well is screened in QA1 and QA2.

When the concentration of ^{14}C and the value of δ^{13}C for the 33 wells are plotted (fig. 10), there is an apparent change in slope and the distribution of modern and pre-modern ground water near 80 pmc. Because a single initial concentration of ^{14}C was not readily indicated from the data, model simulations were run using three values for the initial ^{14}C activity: 80, 85, and 92 pmc. Uncertainty also was present in the ^{14}C activity of the dispersed organic matter present within the aquifer sediments. The concentration of ^{14}C in organic matter present within the peat material obtained from the Upper confining unit (QC1) ranged from roughly 2 to 15 pmc (Deeter, 1979) and provides a likely range of ^{14}C activity present throughout the sediments that make up the hydrogeologic units tapped by the downgradient wells. Multiple simulations were run using ^{14}C concentrations of 2, 10, and 15 pmc.

A matrix showing the estimates of ground-water residence time calculated using different values of the ^{14}C activity in the recharge water and aquifer organic matter is shown in table 7. Estimates of ground-water residence time varied most in the Upper confining unit, where estimates ranged from modern to more than 4,550 years bp. The younger-aged waters were restricted to the upgradient recharge area, whereas the older-aged waters (on the order of thousands of years) were found in both upgradient and downgradient locations.

Many ground waters with shorter residence times were from wells near the area of recharge, and those with longer residence times were from wells either deeper in the ground-water flow system or near the area of ground-water discharge. The spatial distribution is shown in figure 11, which is a composite of ground-water residence times that were estimated near the southern end of SUBASE Bangor. The large residence times of water found in wells tapping the Upper confining unit near Clear Creek drainage (a ground-water discharge zone) are explained by the fact that this ground water previously may have been in deeper parts of the flow system, such as the Sea-level aquifer, and is moving upward as it discharges. Residence times of ground waters in the Sea-level aquifer generally are on the order of thousands of years. However, residence times at several locations were only several hundred years old to modern. The shorter residence times may be explained by intensive pumping at large production wells such as the SUBASE Bangor production well (26N/01E-18P01) that may cause increased downward flow of ground water, resulting in a mixture of modern and pre-modern ground water.

Comparison of Estimates of Ground-Water Residence Times Based on Carbon-14 and Numerical Ground-Water Flow Modeling

The ^{14}C estimates of ground-water residence time were developed to provide an independent constraint that could be used to help calibrate the ground-water flow model developed by van Heeswijk and Smith (2002). At 12 selected wells, van Heeswijk and Smith estimated ground-water residence times using the calibrated flow model with the particle-tracking module MODPATH to trace the paths of 600 hypothetical water particles from a well to the recharge area. The 12 wells were identified as having pre-modern water, based on environmental tracer data. The resulting flow-modeled estimates of ground-water residence time for all particles ranged from 16 to 2,359 years, and the median residence time of the 600 particles modeled for each well ranged from about 100 to 500 years (table 8).

Estimates of ground-water residence time derived from the geochemical and numerical flow modeling were both less than 5,000 years. The minimum residence times estimated using ^{14}C-dating techniques exceeded the median residence times simulated with the flow model for 5 of the 12 simulated sites. Most of the maximum residence times simulated with the flow model were significantly less than the maximum residence times estimated using the ^{14}C-dating techniques. Therefore, the ^{14}C estimates provide an upper limit on the age of ground water in the flow system.

Table 7. Estimated ground-water residence times computed from mass-balance models using a range of initial carbon-14 activity in Vashon aquifer and a range of carbon-14 activity in aquifer organic matter, SUBASE Bangor and vicinity, Kitsap County, Washington

[**Abbreviations:** ^{14}C, carbon-14; pmc, percent modern carbon]

Well No.	Assigned ^{14}C activity in organic matter in aquifer (pmc)	Estimated ground-water residence time for assigned initial ^{14}C activity (years)			Range of estimated ground-water residence time (years)
		92 pmc	85 pmc	80 pmc	
Upper confining unit (QC1)					
26N/01E-17C01	Model does not utilize organic C	2,990	2,350	1,840	1,840 to 2,990
27N/01E-35C01	15	3,080	2,440	1,940	1,900 to 3,080
	10	3,070	2,430	1,920	
	2	3,050	2,410	1,900	
Permeable interbeds within Upper confining unit (QC1pi)					
25N/01E-07J02	15	510	41	-330	50 to 510
	10	-310	-830	-1,240	
	2	-1,610	-2,220	-2,710	
25N/01E-08J02	15	1,700	1,180	760	50 to 1,700
	10	1,210	650	210	
	2	350	-270	-770	
25N/01E-08Q03	15	4,550	4,110	3,760	920 to 4,550
	10	3,550	3,060	2,670	
	2	2,010	1,400	920	
25N/01E-09N02	15	4,500	3,970	3,560	2,130 to 4,500
	10	4,040	3,480	3,040	
	2	3,250	2,630	2,130	
27N/01E-22Q04	15	-975	-1,570	-2,040	All models result in futuristic dates and are invalid for estimating ^{14}C age of ground water
	10	-1,170	-1,780	-2,270	
	2	-1,490	-2,130	-2,630	
Sea-level aquifer (QA1)					
25N/01E-06D04	15	2,480	1,940	1,500	330 to 2,480
	10	2,110	1,540	1,080	
	2	1,470	850	330	
25N/01W-01B02	15	4,550	4,000	3,540	2,750 to 4,550
	10	4,550	3,720	3,240	
	2	3,890	3,250	2,750	
27N/01E-22Q05	15	1,480	850	340	120 to 1,480
	10	1,420	790	280	
	2	1,330	690	120	
26N/01E-32L05	15	1,040	400	-100	50 to 1,040
	10	1,010	370	-140	
	2	960	320	-190	
26N/01E-36R03	15	450	-180	-680	50 to 450
	10	410	-220	-730	
	2	340	-300	-810	
Deep aquifer (QA2)					
26N/01E-31R01[1]	15	2,120	1,540	1,090	180 to 2,120
	10	1,820	1,220	750	
	2	1,320	690	180	
26N/01E-32L04	15	1,510	870	370	250 to 1,510
	10	1,490	860	350	
	2	1,420	780	250	

[1]Well is screened in QA1 and QA2.
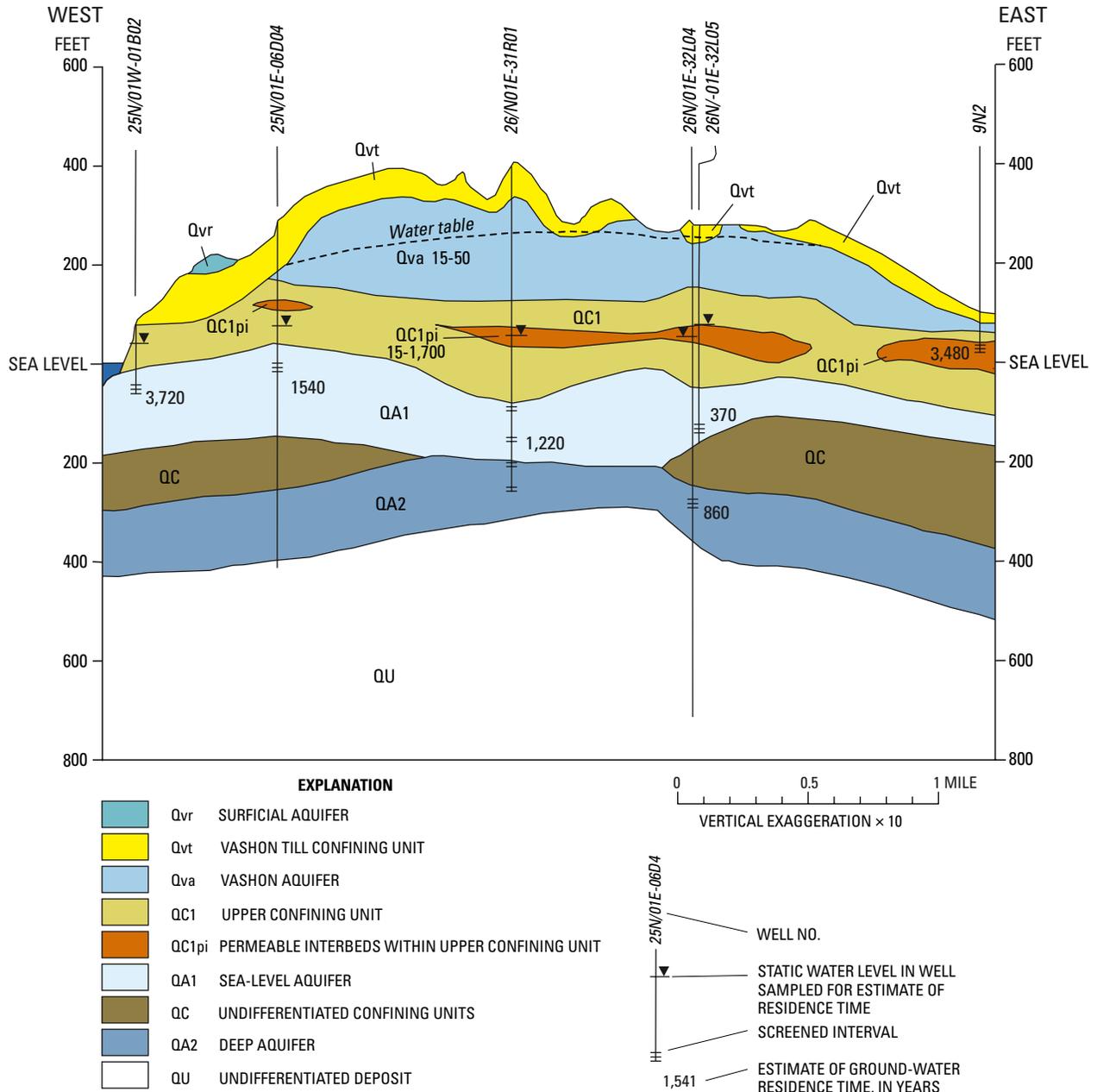
Figure 11. Distribution of estimated ground-water residence time in the ground-water system beneath SUBASE Bangor and vicinity, Kitsap County, Washington.

Individual estimates are based on initial ^{14}C activity of 85 percent modern carbon (pmc) and ^{14}C activity of aquifer organic matter of 10 pmc (see table 7). Section based on hydrogeologic section F-F' of Kahle (1998, pl. 3). Ground-water residence times for Qva and QC1pi projected from other locations.

Table 8. Comparison of estimates of ground-water residence time (age) based on the concentration of carbon-14 and numerical simulation of ground-water flow, SUBASE Bangor and vicinity, Kitsap County, Washington

[Modified from van Heeswijk and Smith (2002)]

| Well No. | Estimate of ground water residence time, in years | | | |
| | Range based on [14]C | Based on numerical ground-water flow modeling | | |
		Median	Minimum	Maximum
Upper confining unit (QC1)				
27N/01E-35C01	1,900-3,080	104	41	168
Permeable interbeds within the Upper confining unit (QC1pi)				
25N/01E-07J02	50-510	163	106	199
25N/01E-08J02	50-1,700	219	175	361
25N/01E-08Q03	920-4,550	235	175	407
25N/01E-09N02	2,130-4,500	237	190	359
Sea-level aquifer (QA1)				
25N/01E-06D04	330-2,480	103	32	212
25N/01W-01B02	2,750-4,550	101	54	160
26N/01E-32L05	50-1,040	211	16	428
26N/01E-36R03	50-450	97	71	392
27N/01E-22Q05	120-1,480	399	242	2,359
Deep aquifer (QA2)				
26N/01E-31R01[1]	180-2,120	296	182	661
26N/01E-32L04	250-1,510	478	360	1,816

[1]Well is screened in QA1 and QA2.

The residence times simulated with the ground-water flow model were consistent with tritium and CFC concentration data—the median simulated residence times were all greater than 50 years. The minimum residence times simulated for some particles arriving at wells 25N/01E-06D04 and 26N/01E-32L05 were 32 and 16 years, respectively, indicating that some modern water may be present in these wells. However, the minimum flow-modeled estimates may represent only a small percentage of the water in each well, hence the presence of modern environmental tracers would likely be small and may not be detectable.

The discrepancies between the ^{14}C and ground-water flow model estimates of ground-water residence time likely resulted from multiple factors, including uncertainties related to geochemical processes and measurements, as well as uncertainties related to the flow-model representation of the hydrologic system. Uncertainties in the numerical simulation of ground-water flow include the conceptualization of the ground-water flow system, vertical and horizontal discretization of the model grid, simulated aquifer and confining unit properties such as porosities and vertical and horizontal hydraulic conductivities, and simulated ground-water recharge. In their simulation, van Heeswijk and Smith (2002) used values for effective porosity of till = 0.15; sand and gravel = 0.3; and clay and silt = 0.2 that they considered to be the largest appropriate for the hydrogeologic units of the Bangor area and that increased their estimated of ground-water residence time. It is possible that some of the flow paths simulated by the particle-tracking software do not accurately represent the actual flow paths of water particles that were sampled. This may explain large discrepancies between flow-modeled and ^{14}C-modeled ages estimated for wells 25N/01W-01B02 and 25N/01E-09N02, which are located near regional discharge locations. Within the flow model, the particle paths to these wells may have originated from nearby recharge areas rather than following deeper regional

ground-water flow paths that are indicated by the evolved ground-water quality observed in the samples from these wells. Additionally, near-stagnant or low-velocity parts of the flow system may not be adequately simulated by the numerical flow model and may explain the discrepancy between the flow-modeled and ^{14}C-modeled ages in well 25N/01E-08Q03. The limitations of the ground-water flow model are discussed in more detail by van Heeswijk and Smith (2002). Uncertainty in the isotope data can result in large variability in ^{14}C model results, as demonstrated by the data in this report ().

In their simulation, van Heeswijk and Smith (2002) used values for effective porosity of till = 0.15; sand and gravel = 0.3; and clay and silt = 0.2 that they considered to be the largest appropriate for the hydrogeologic units of the Bangor area and that resulted in upper limits of residence times that could be simulated with their numerical flow model. Despite simulating upper limits of residence times, the model-simulated values generally are smaller than the ^{14}C values. It is possible that some of the flow paths simulated by the particle-tracking software do not accurately represent the actual flow paths of water particles that were sampled, particularly near regional ground-water discharge areas where ground water flow lines converge. This may explain large discrepancies between flow-modeled and ^{14}C-modeled ages estimated for wells 25N/01W-01B02 and 25N/01E-09N02, which are located near regional discharge locations. Within the flow model, the particle paths to these wells may have originated from nearby recharge areas rather than following deeper regional ground-water flow paths that are indicated by the evolved ground-water quality observed in the samples from these wells. Additionally, near-stagnant or low-velocity parts of the flow system may not be adequately simulated by the numerical flow model and may explain the discrepancy between the flow-modeled and ^{14}C-modeled ages observed in well 25N/01E-8Q03. The limitations of the ground-water flow model are discussed in more detail by van Heeswijk and Smith (2002).

Uncertainty in the ^{14}C estimates of ground-water residence time are often on the order of 1,000 years, in large part due to uncertainty in the estimate of the initial ^{14}C activity of recently recharged ground water (Kalin, 2000). Information about the mineralogy of the aquifer sediments in the Bangor area is very limited and undoubtedly incomplete. Thus, the selection of phases used in the NETPATH model might be improved with additional information. Likewise, NETPATH was only able to model a limited number of phases, and it is likely that in glacial sediments with heterogeneous source areas there are more than seven chemically reactive phases in the ground-water system. Diffusion in ground-water systems that contain substantial confining units also may reduce the concentration of ^{14}C in ground water, resulting in a bias toward larger estimates of ground-water residence time, as suggested by Sanford (1997) and Bethke and Johnson (2002). In ground-water systems that contain many confining units, recently recharged ground water containing large concentrations of ^{14}C may leave the aquifer by diffusion into adjacent confining units; likewise, at downgradient locations ground water that is depleted in ^{14}C may defuse into the aquifer. Both processes would diminish the ^{14}C content of ground water, resulting in an over-estimate of ground-water residence times.

Although differences between estimates of ground-water residence times based on the geochemical models of ^{14}C and numerical simulation of ground-water flow can be large, the results do confirm that travel times to the stratigraphically deeper sections of the aquifer system on and near SUBASE Bangor are on the order of hundreds to thousands of years. The use of environmental tracers provides additional information in instances where there is rapid movement of ground water to stratigraphically deeper units along routes that bypass portions of the aquifer system, such as in well 27N/01E-27J02.

SUMMARY

The U.S. Geological Survey, in cooperation with the U.S. Navy, estimated the residence time of ground water at different locations within the ground-water system in the vicinity of Naval Submarine Base (SUBASE) Bangor. Water samples were collected from 33 wells for analysis of selected environmental tracers, inorganic constituents, and carbon species. Estimates of ground-water residence time were based on concentrations of ^{14}C, tritium, and chlorofluorocarbons. This work is part of a larger study to understand the regional ground-water flow system at SUBASE Bangor and vicinity.

The ground-water system is composed of alternating coarse and fine-grained deposits, mostly of glacial and inter-glacial origin. The shallower aquifer (Vashon aquifer) is composed of glacial outwash sand and is beneath a discontinuous surficial till deposit. The deeper Sea-level and Deep aquifers are fluvial sand and gravel deposit separated from the shallow aquifer by a thick layer of fine-grained interglacial material that in places contain lenses or interbeds of coarse material that yield usable quantities of ground water.

The concentration of carbon increased in ground water along ground-water flow paths, indicating that additional carbon was being incorporated into solution from the aquifer matrix. The carbon present within the aquifer material will affect the ^{14}C activity of ground water and thus affect the estimate of ground-water residence time. Carbon-13 ratios indicate that the source of the additional carbon is dispersed organic carbon present primarily in the confining unit separating the two aquifers. Dissolved methane was present in some ground water at concentrations larger than atmospheric concentrations in more than one-half of the samples analyzed. The source of the methane is the microbial decomposition of organic matter. The additional carbon in the form of methane reduces the activity of ^{14}C in ground water, resulting in uncorrected estimates of residence time that are substantially larger than the actual ground-water residence time.

Geochemical modeling of the mass transfer of carbon and carbon isotopes, using the NETPATH code, was used to estimate ground-water residence times based on ^{14}C concentrations. The modeling also accounted for the interaction of carbon in solution with carbon present in the aquifer material. The overall reactions that were modeled were the oxidation of organic matter with the production of methane, the dissolution of silicate minerals and the precipitation of goethite, and the ion exchange of calcium for sodium. Ground-water residence times based on ^{14}C were estimated for 14 locations in the ground-water flow system and ranged from modern to about 4,550 years before present.

Tritium concentrations were used to detect the presence of modern ground water and to identify water samples that were a mixture of modern and pre-modern ground water. Three of the samples analyzed contained both modern and pre-modern waters. Mixed ground water is believed to result from large pumping stresses that may have increased downward movement of younger water into lower aquifers, resulting in a mixture of relatively young and old waters.

The shortest residence times are found largely near the area of recharge, whereas longer residence times are in water from wells either deeper in the ground-water flow system or near the area of ground-water discharge.

At selected wells where estimates of ground-water residence times also had been estimated by particle-tracking analysis using a ground-water flow model, estimates of the residence time of ground water based on the analysis of ^{14}C were often larger. At upgradient locations close to recharge areas, estimates of residence time based on analysis of ^{14}C were in the range of 50 to 500 years. However, for wells located near regional ground-water discharge areas, estimates of ground-water residence time based on analysis of ^{14}C were often in the range of 2,000 to 4,500 years and were as much as 10 times larger or more than estimates derived from a ground-water flow model.

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Appendix A. Quality-assurance data for isotope analysis of ground-water samples from wells on SUBASE Bangor and vicinity, Kitsap County, Washington

[Data as reported by analyzing laboratory and not rounded. **NH₄-SrCl₂**: 50 milliliters added to sample in the file prior to sending to laboratory. **Water**: Water only sent to laboratory. **Powder**: Powder precipitate of $SrCO_3$ prepared in Reston laboratory and then sent to analyzing laboratory. **AMS**, Accelerator Mass Spectrometer; **MS**, mass spectrometer. $\delta^{13}C$, delta carbon-13; pCi/L, picoCuries per liter; NH_4-$SrCl_2$, ammoniacal-strontium-chloride. —, no data]

Well No.	Carbon-14 AMS			Carbon-13 AMS			Carbon-13 MS
	NH_4-$SrCl_2$	Water	Powder	NH_4-$SrCl_2$	Water	Powder	
	(percent modern carbon)			($\delta^{13}C$, per mil)			
25N/01E-06D04	–	40.81±0.32	–	–	-19.3	–	-19.2
	–	39.94±0.32	–	–	-18.6	–	-18.9
25N/01E-07J01	117.18±0.75	–	–	-21.09	–	–	–
25N/01E-07N02D1	68.72±0.48	63.03±0.47	66.97±0.46	-21.64	-22.44	-22.05	-21.76
25N/01E-12J01	–	66.97±0.52	70.15±0.46	-21.60	-21.31	-21.69	-21.37
26N/01E-17C01	–	59.06±0.44	60.47±0.39	–	-18.06	-17.64	-17.47
26N/01E-18P06	83.99±0.58	78.63±0.57	82.01±0.50	-21.75	-20.57	-21.41	-21.19
26N/01E-30L01	68.79±0.52	66.99±0.49	66.15±0.57	-21.75	-21.80	-21.58	-21.23
	70.39±0.50	–	–	-21.10	–	–	–
26N/01E-32M01	116.15±0.74	–	–	-24.1	–	–	–
27N/01E-35C01	61.62±0.48	–	–	-18.64	–	–	–
	62.02±0.46	–	–	-18.92	–	–	–

Well No.	Tritium (pCi/L)	Deuterium (δ^2H, per mil)	Oxygen-18 ($\delta^{18}O$, per mil)
25N/01E-06D04	<0.3±0.6	-79.20	-11.20
	0.4±0.6	-79.10	-11.27
25N/01E-07J01	15.5±1.1	-74.9	-10.53
	16.6±1.1	–	–
25N/01E-07N02D1	18.0±1.2	-76.5	-10.91
25N/01E-12J01	27.0±1.2	-76.4	-10.99
26N/01E-17C01	<0.3±0.6	-79.0	-11.25
26N/01E-18P06	24.0±1.6	-80.3	-11.28
26N/01E-30L01	12.0±0.08	-79.4	-11.23
26N/01E-32M01	20.4±1.3	-78.2	-10.81
	21.4±1.4	–	–
27N/01E-35C01	0.7±0.6	-78.9	-11.21

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S.E. Cox

Estimates of Residence Time and Related Variations in Quality of Ground Water Beneath Submarine Base Bangor and Vicinity, Kitsap County, Washington