National Water-Quality Assessment Program

Chloride in Groundwater and Surface Water in Areas Underlain by the Glacial Aquifer System, Northern United States

Scientific Investigations Report 2009–5086

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

Cover. Rock salt being unloaded off of a ship in Oswego, New York, October, 2007
(James Rajotte/The New York Times/Redux)

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Foreword

The U.S. Geological Survey (USGS) is committed to providing the Nation with credible scientific information that helps to enhance and protect the overall quality of life and that facilitates effective management of water, biological, energy, and mineral resources (*http://www.usgs.gov/*). Information on the Nation's water resources is critical to ensuring long-term availability of water that is safe for drinking and recreation and is suitable for industry, irrigation, and fish and wildlife. Population growth and increasing demands for water make the availability of that water, now measured in terms of quantity and quality, even more essential to the long-term sustainability of our communities and ecosystems.

The USGS implemented the National Water-Quality Assessment (NAWQA) Program in 1991 to support national, regional, State, and local information needs and decisions related to water-quality management and policy (*http://water.usgs.gov/nawqa*). The NAWQA Program is designed to answer: What is the condition of our Nation's streams and groundwater? How are conditions changing over time? How do natural features and human activities affect the quality of streams and groundwater, and where are those effects most pronounced? By combining information on water chemistry, physical characteristics, stream habitat, and aquatic life, the NAWQA Program aims to provide science-based insights for current and emerging water issues and priorities. From 1991-2001, the NAWQA Program completed interdisciplinary assessments and established a baseline understanding of water-quality conditions in 51 of the Nation's river basins and aquifers, referred to as Study Units (*http://water.usgs.gov/nawqa/studyu.html*).

In the second decade of the Program (2001–2012), a major focus is on regional assessments of water-quality conditions and trends. These regional assessments are based on major river basins and principal aquifers, which encompass larger regions of the country than the Study Units. Regional assessments extend the findings in the Study Units by filling critical gaps in characterizing the quality of surface water and groundwater, and by determining status and trends at sites that have been consistently monitored for more than a decade. In addition, the regional assessments continue to build an understanding of how natural features and human activities affect water quality. Many of the regional assessments employ modeling and other scientific tools, developed on the basis of data collected at individual sites, to help extend knowledge of water quality to unmonitored, yet comparable areas within the regions. The models thereby enhance the value of our existing data and our understanding of the hydrologic system. In addition, the models are useful in evaluating various resource-management scenarios and in predicting how our actions, such as reducing or managing nonpoint and point sources of contamination, land conversion, and altering flow and (or) pumping regimes, are likely to affect water conditions within a region.

Other activities planned during the second decade include continuing national syntheses of information on pesticides, volatile organic compounds (VOCs), nutrients, selected trace elements, and aquatic ecology; and continuing national topical studies on the fate of agricultural chemicals, effects of urbanization on stream ecosystems, bioaccumulation of mercury in stream ecosystems, effects of nutrient enrichment on stream ecosystems, and transport of contaminants to public-supply wells.

The USGS aims to disseminate credible, timely, and relevant science information to address practical and effective water-resource management and strategies that protect and restore water quality. We hope this NAWQA publication will provide you with insights and information to meet your needs, and will foster increased citizen awareness and involvement in the protection and restoration of our Nation's waters.

The USGS recognizes that a national assessment by a single program cannot address all water-resource issues of interest. External coordination at all levels is critical for cost-effective management, regulation, and conservation of our Nation's water resources. The NAWQA Program, therefore, depends on advice and information from other agencies—Federal, State, regional, interstate, Tribal, and local—as well as nongovernmental organizations, industry, academia, and other stakeholder groups. Your assistance and suggestions are greatly appreciated.

Matthew C. Larsen

Associate Director for Water

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Conversion Factors

Multiply	By	To obtain
	Length	
inch (in.)	2.54	centimeter (cm)
inch (in.)	25.4	millimeter (mm)
foot (ft)	0.3048	meter (m)
mile (mi)	1.609	kilometer (km)
	Area	
square mile (mi^2)	259.0	hectare (ha)
square mile (mi^2)	2.590	square kilometer (km^2)
	Volume	
gallon (gal)	3.785	liter (L)
gallon (gal)	0.003785	cubic meter (m^3)
gallon (gal)	3.785	cubic decimeter (dm^3)
million gallons (Mgal)	3,785	cubic meter (m^3)
cubic foot (ft^3)	28.32	cubic decimeter (dm^3)
cubic foot (ft^3)	0.02832	cubic meter (m^3)
	Flow rate	
cubic foot per second (ft^3/s)	0.02832	cubic meter per second (m^3/s)
million gallons per day (Mgal/d)	0.04381	cubic meter per second (km^3/s)
inch per year (in/yr)	25.4	millimeter per year (mm/yr)
	Mass	
ounce, avoirdupois (oz)	28.35	gram (g)
pound, avoirdupois (lb)	0.4536	kilogram (kg)
ton, short (2,000 lb)	0.9072	megagram (Mg)
ton per year (ton/yr)	0.9072	megagram per year (Mg/yr)
ton per year (ton/yr)	0.9072	metric ton per year
tons per square mile (ton/mi^2)	350.1947	kilograms per square kilometer (kg/km^2)
pound (lb)	453,592.4	milligram (mg)
pounds per day (lb/d)	453,592.4	milligram per day (mg/d)
pound (lb)	0.4535924	kilogram (kg)

Concentrations of chemical constituents in water are given in milligrams per liter (mg/L).

Horizontal coordinate information is referenced to the North American Datum of 1983 (NAD 83).

Chloride in Groundwater and Surface Water in Areas Underlain by the Glacial Aquifer System, Northern United States

By John R. Mullaney, David L. Lorenz, and Alan D. Arntson

Abstract

A study of chloride in groundwater and surface water was conducted for the glacial aquifer system of the northern United States in forested, agricultural, and urban areas by analyzing data collected for the National Water-Quality Assessment Program from 1991 to 2004.

Groundwater-quality data from a sampling of 1,329 wells in 19 states were analyzed. Chloride concentrations were greater than the secondary maximum contaminant level established by the U.S. Environmental Protection Agency of 250 milligrams per liter in 2.5 percent of samples from 797 shallow monitoring wells and in 1.7 percent of samples from 532 drinking-water supply wells. Water samples from shallow monitoring wells in urban areas had the largest concentration of chloride, followed by water samples from agricultural and forested areas (medians of 46, 12, and 2.9 milligrams per liter, respectively).

An analysis of chloride:bromide ratios, by mass, and chloride concentrations compared to binary mixing curves for dilute groundwater, halite, sewage and animal waste, potassium chloride fertilizer, basin brines, seawater, and landfill leachate in samples from monitoring wells indicated multiple sources of chloride in samples from wells in urban areas and agricultural areas. Water from shallow monitoring wells in urban areas had the largest chloride:bromide ratio, and samples with chloride:bromide ratios greater than 1,000 and chloride concentrations greater than 100 milligrams per liter were dominated by halite; however, the samples commonly contained mixtures that indicated input from sewage or animal waste. Chloride:bromide ratios were significantly larger in samples from public-supply drinking-water wells than from private drinking-water wells, and ratios were significantly larger in all drinking-water wells in eastern and central regions of the glacial aquifer system than in west-central and western regions of the glacial aquifer system.

Surface-water-quality data collected regularly during varying time periods from 1991–2004 from 100 basins dominated by forested, agricultural, or urban land in 15 states were analyzed to determine maximum measured chloride concentrations. Samples from 15 sites in east, central, and west-central areas, collected primarily in winter, had chloride concentrations higher than the U.S. Environmental Protection Agency recommended chronic criterion concentration for aquatic life of 230 milligrams per liter. Concentrations of chloride in base-flow samples were predictive of maximum measured chloride concentrations, indicating that inputs of chloride from groundwater and (or) point-source wastewater discharges increase the likelihood of samples exceeding the recommended chronic aquatic criterion. Multiple linear regression analyses showed that the density of major roads, potential evapotranspiration, and the percentage of annual runoff from saturated overland flow were significant factors in describing the range of maximum measured chloride concentrations in the basins studied.

Chloride loads and yields were determined at 95 surface-water-monitoring stations in basins dominated by forested, agricultural, or urban land. Annual chloride yield was largest in the urban basins (median of 88 tons per square mile) and smallest in the forested basins (median of 6.4 tons per square mile). The median chloride yield in the agricultural basins was 15.4 tons per square mile. Multiple linear regression analyses showed that the density of highways (roads in U.S. highway system), the number of major wastewater discharges in the basin, potential evapotranspiration, and urban minus agricultural land area were significant factors in describing the range of average annual chloride yields.

Upward trends in chloride loads were apparent in several urban basins for which additional long-term data were available. Increases in chloride loads over time may be related to a variety of factors, including increases in road area and consequent deicing, increases in wastewater and septic-system discharges, recycling of chloride from drinking water, and leachate from landfills and salt storage areas.

Introduction

The use of salt has increased measurably in the United States since 1950 (Kostick, 1993; Kostick and others, 2007) (fig. 1A), and the major increase in the use of salt has been for deicing of roads, parking lots, and other impervious surfaces during the winter months (fig. 1B). The application of salt for deicing these surfaces has raised awareness of potential adverse effects on water resources (Bubeck and others, 1971; Huling and Hollocher, 1972; Wulkowicz and Saleem, 1974).

Widespread upward trends in chloride concentrations in streams nationwide have been reported from 1974–81 (Smith and others, 1987). Similar trends have been reported in Connecticut from the 1970s to the 1990s (Trench, 1996; Colombo and Trench, 2002) and in New Jersey (Hay and Campbell, 1990; Robinson and others, 1996). Several studies have shown elevated concentrations of chloride and sodium in glacial aquifers related to urban land use (Grady and Mullaney, 1998; Fong, 2000; Thomas, 2000; Savoca and others, 2000).

The primary concern for water quality is the degradation of groundwater and surface water that may be used for drinking-water supply or for aquatic habitat. The U.S. Environmental Protection Agency (USEPA) has set a secondary maximum contaminant level (SMCL) of 250 mg/L for chloride in drinking water (U.S. Environmental Protection Agency, 1992). The SMCL for chloride is an unenforceable guideline that relates to the aesthetics of the water and the perceived salty taste of water at concentrations above 250 mg/L. The USEPA-recommended chronic criterion for aquatic life is a 4-day average chloride concentration of 230 mg/L with an occurrence interval of once every 3 years, and the recommended acute criterion concentration for chloride is 860 mg/L (U.S. Environmental Protection Agency, 1988). The acute criterion relates to a 1-hour average concentration with a recurrence interval of less than once every 3 years. Other concerns regarding salt inputs include the effects of cation-exchange reactions on the quality of water (Granato and others, 1995).

The U.S. Geological Survey (USGS) has been collecting data since 1991 on the quality of water in the Nation’s aquifers and streams through the National Water-Quality Assessment (NAWQA) Program. Data are synthesized at the scale of regional principal aquifers (Lapham and others, 2005) to understand water-quality issues affecting aquifers used commonly for domestic and public water supply. The glacial aquifer system (Warner and Arnold, 2005) is the largest principal aquifer (fig. 2) in the United States, in terms of areal extent and its use as a source for drinking water. The glacial aquifer system is considered here to include all unconsolidated aquifers overlying bedrock north of the line of continental glaciation. Warner and Arnold (2005) divided the glaciated area in the United States into four major glacial regions (fig. 2) based on differing glacial source material: east, central, west-central and west.

The glacial aquifer system also is a source of surface water, because the aquifer and stream systems are commonly in hydraulic connection. Therefore, the discharge of groundwater to streams affects the quality of surface water, and surface water at times can become a source of groundwater.

Purpose and Scope

This report describes the concentrations of salt-related constituents (primarily chloride) in shallow groundwater and surface water in the glaciated northern United States, on the basis of water-quality data collected from 1991 to 2004 for the USGS NAWQA Program at 1,329 wells in 19 states and 100 surface-water sites in 15 states. The report presents information on natural and anthropogenic sources of chloride in the environment, concentrations of sodium and chloride in groundwater, concentrations of chloride in surface water, and estimates of chloride loads and yields from selected basins in the study area. The report also provides information on the relation between ancillary variables (land-use, land-cover, hydrologic, and climatic variables) and the concentrations and yields of chloride in surface water.

Groundwater data analyzed for this study generally were collected from coarse-grained glacial aquifers, and therefore the results may not translate to other areas in the glacial aquifer where coarse-grained deposits are absent, and the surficial material is glacial till or bedrock.

Sources of Salts to Water Resources

Natural Sources

Natural sources of salts to water resources include (1) the oceans; (2) the natural weathering of bedrock, surficial materials, and soils; (3) geologic deposits containing halite, or saline groundwater (brines); and (4) volcanic activity.

The oceans typically contain about 19,000 milligrams per liter (mg/L) of chloride (Feth, 1981). Sodium and chloride are the most abundant ions present in the atmosphere above the oceans, and atmospheric deposition of chloride and sodium from the oceans is highest along coastal areas. Atmospheric deposition may contain natural sources of chloride from the oceans, and dusts from surficial salt deposits such as in playa lakes, as well as chloride from anthropogenic sources, such as manufacturing and power generation.

Atmospheric Deposition

From 1994 to 2003, wet deposition of chloride contributed an average of 0.04 to 6.2 ton/mi^2 to the glacial aquifer system study area (table 1, fig. 3) based on the data from the National Atmospheric Deposition Program (NADP) (2006). The largest amount of wet deposition of chloride was on the eastern and western coastal parts of the study area, including the Puget Sound area and the New England coastal basins. The

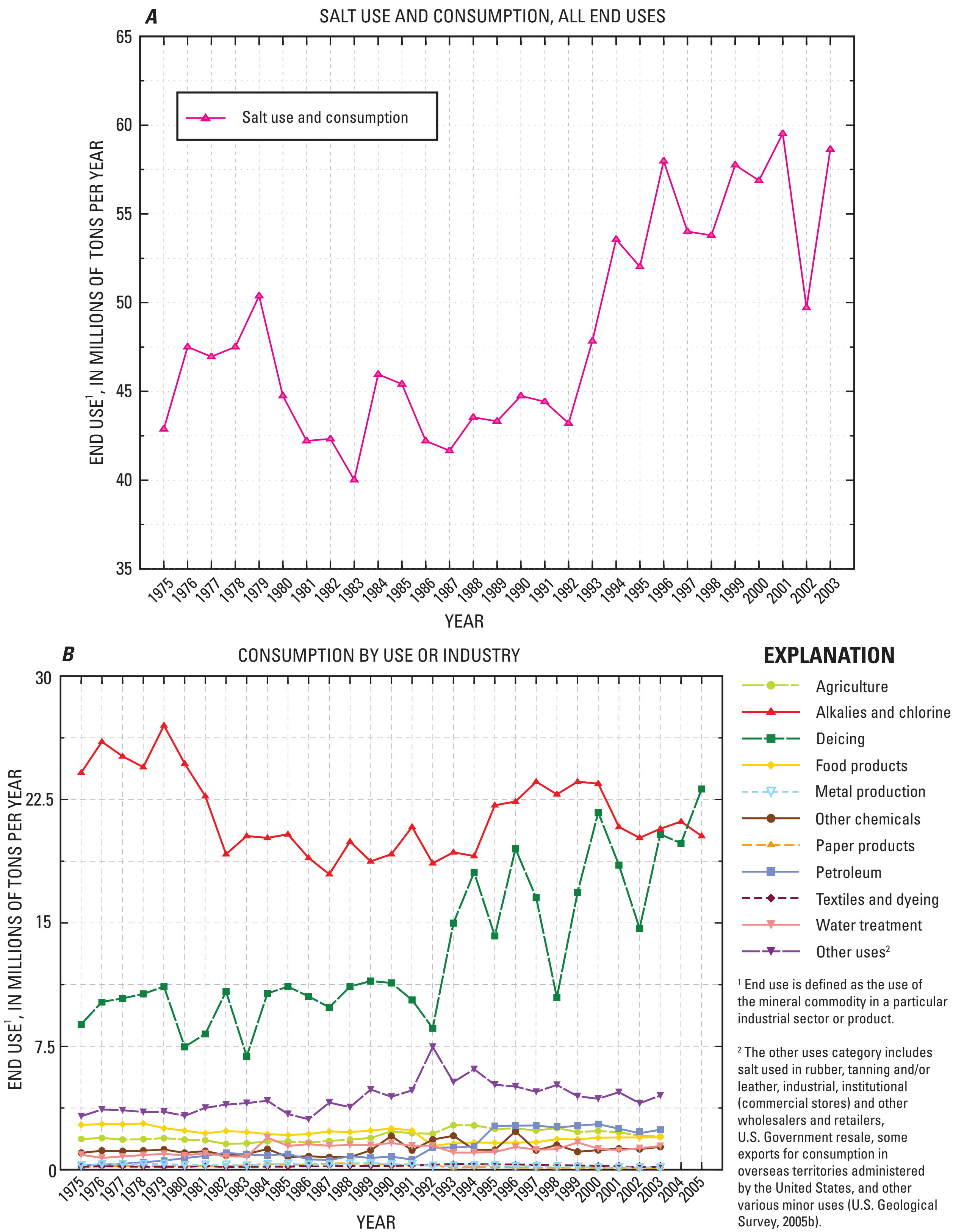


Figure 1. Salt use and consumption in the United States by *(A)* all end users, 1975–2003 (data from U.S. Geological Survey, 2005b), and *(B)* consumption by use or industry, 1975–2005 (data from U.S. Geological Survey, 2005b; Kostick and others, 2007).

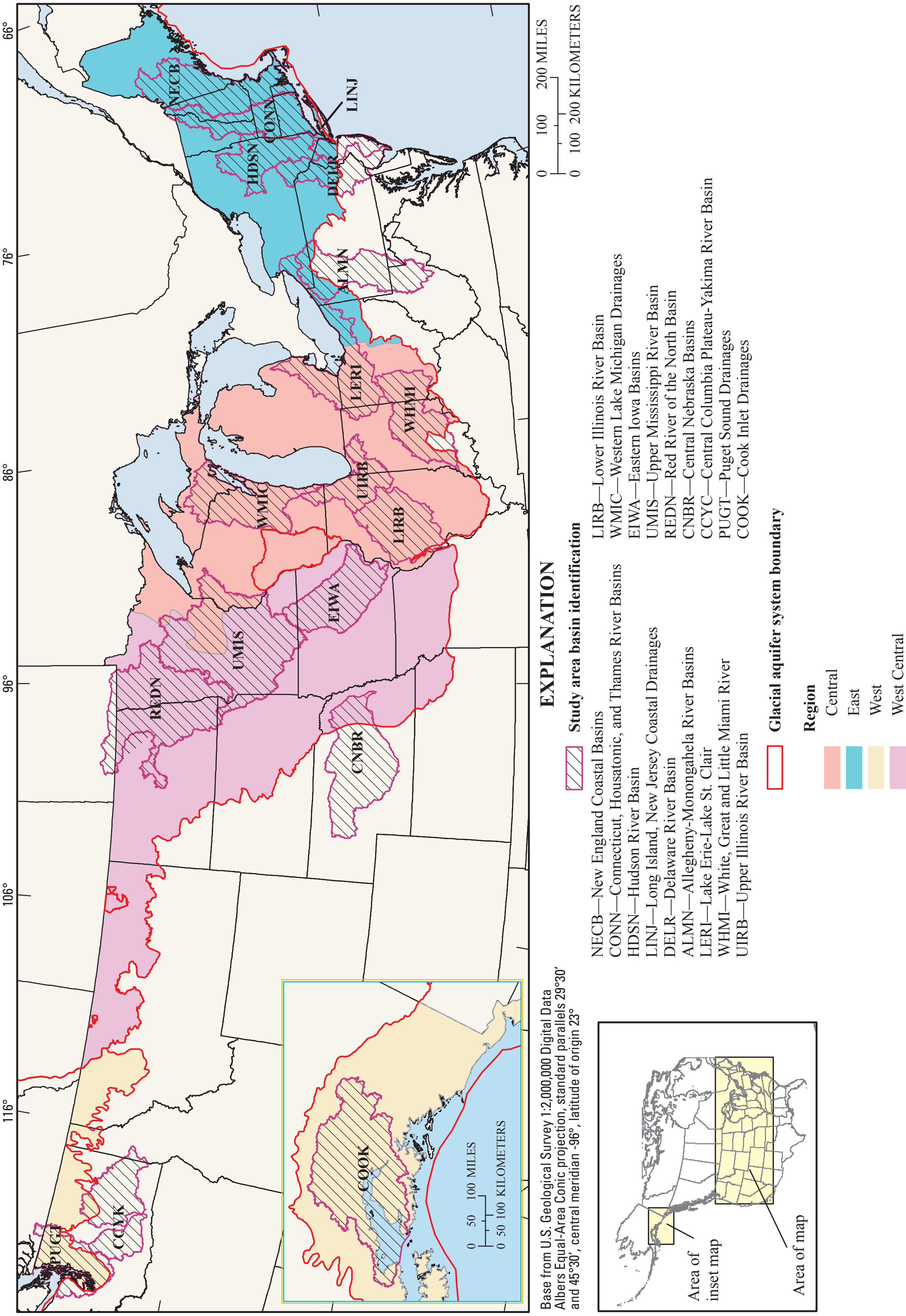


Figure 2. Glacial aquifer system and National Water-Quality Assessment Program study basins, northern United States.

Table 1. Estimated wet deposition of chloride on the glacial aquifer system, northern United States, 1994–2003.

[Values are in tons per square mile per year. Data analyzed from the National Atmospheric Deposition Program, 2006]

Region	Minimum	Maximum	Mean
East	0.19	4.16	0.71
Central	.09	.64	.25
West central	.04	.40	.13
West	.06	6.22	.91

lowest amount of wet deposition of chloride was in the central parts of the study area, including Iowa, Minnesota, North Dakota, and South Dakota. In basins in the United States with little or no human activity, the yield of chloride from streams is correlated with the amount of precipitation. The chloride yield from atmospheric deposition is related to precipitation quantity and has been reported to contribute on average 28 to 62 percent of the chloride yield in undeveloped basins throughout the United States. Basins with crystalline bedrock were associated with the largest percentage of atmospheric chloride yield, followed, in order, by basins underlain by sandstone and basins underlain by limestone (Peters, 1984).

Chloride concentrations in streams and groundwater that originated from wet deposition of chloride are different from concentrations measured in precipitation because of the concentration of chloride by the process of evapotranspiration. The average concentrations of chloride from wet deposition in runoff (surface and groundwater) can be calculated as the wet deposition of chloride divided by the runoff (fig. 4). This calculation indicates that average concentrations in runoff attributable to wet deposition are typically 0.1–2.0 mg/L.

Weathering of Common Rocks, Minerals, and Soils

In addition to the chloride deposited on a basin by precipitation, there is a natural input from the weathering of rocks and minerals. Chlorine is present in several minerals in common rocks, and its release to waters as chloride ions is generally slow and through processes other than dissolution (Feth, 1981). The chloride in excess of the load from precipitation was attributed to the weathering of the mineral hornblende in glacial tills in two Adirondack Mountain basins in New York (Peters, 1991).

The chloride yields from 21 basins with no permanent residents, underlain by limestone, sandstone, and crystalline rocks, were analyzed by Peters (1984), who concluded, on the basis of a regression analysis, that 77 percent of the variation in chloride yield from these basins could be explained by the quantity of precipitation a basin receives. The relation to precipitation probably includes direct wet deposition of chloride, as well as chemical weathering, which increases with precipitation. A subset of 10 of these basins[1] with very low population density was identified in the glacial aquifer system. The difference between the chloride yield attributable to direct input from precipitation and the amount attributable to the relation between precipitation and chloride yield probably represents an estimate of the amount derived from chemical weathering. This difference ranged from 0 to 72 percent of the chloride yield. Chloride yields from these 10 basins ranged from 0.7 to 68 tons/mi^2, although all but one site had yields less than or equal to 9 tons/mi^2. The estimated annual load from chemical weathering in these 10 basins, on the basis of the above percentage, ranged from 0 to 2 tons/mi^2 with an average of 0.9 tons/mi^2 (Peters, 1984).

Salt Deposits and Brines

Bedded salt deposits underlie parts of the study area, including parts of Michigan, Western New York, Ohio, and Pennsylvania (Norris, 1978). New York, Ohio, and Michigan produce much of the rock salt and other salts derived from brines in the United States. Most halite deposits occur at depth or at the downgradient ends of groundwater systems (Feth, 1981) and therefore generally may not affect the shallow-groundwater quality of the glacial aquifer system. Water with high concentrations of dissolved solids is present in sedimentary bedrock at depth in many parts of the study area (Feth, 1965). These halite deposits and brines may be a salinity source for groundwater in zones of flow convergence, where water of different ages and from different aquifers (surficial to deep bedrock) have mixed. Therefore, it is possible that part of the chloride budget for some basins in these areas may be from dissolution of halite deposits or mixing with brines. This component of the chloride budget is evidenced by saline springs in parts of the study area, such as Michigan, Illinois, and New York (Michigan Department of Environmental Quality, 1994; Panno and others, 2006; Rao and others, 2005).

Anthropogenic Sources

The use of salt in the United States has increased from 42.9 million tons in 1975 to nearly 58.5 million tons in 2005 (Kostick and others, 2007) (fig. 1A). The largest use of salt has been in the chloralkali industry that produces chlorine and sodium hydroxide (Kostick, 1993) (fig. 1B). In 2005, salt used by the chloralkali industry represented 34.7 percent of the end use of salt. The second largest use of salt since 1975 has been deicing. Deicing use has increased since then, and was greater than that for the chloralkali industry in 2005, when it represented 39.5 percent of the end use of salt in the United States (Kostick and others, 2007). Other uses of salt (totaling about 25 percent of end use in 2005) include agriculture, food processing, metal processing, paper production, textiles and dyeing, petroleum production, water treatment, and other

[1] Basins included the following stations from Peters (1984), table 1: 4045500, 4057004, 4126520, 4132052, 6452000, 6478500, 1066000, 4014500, 5124480, 12447390.

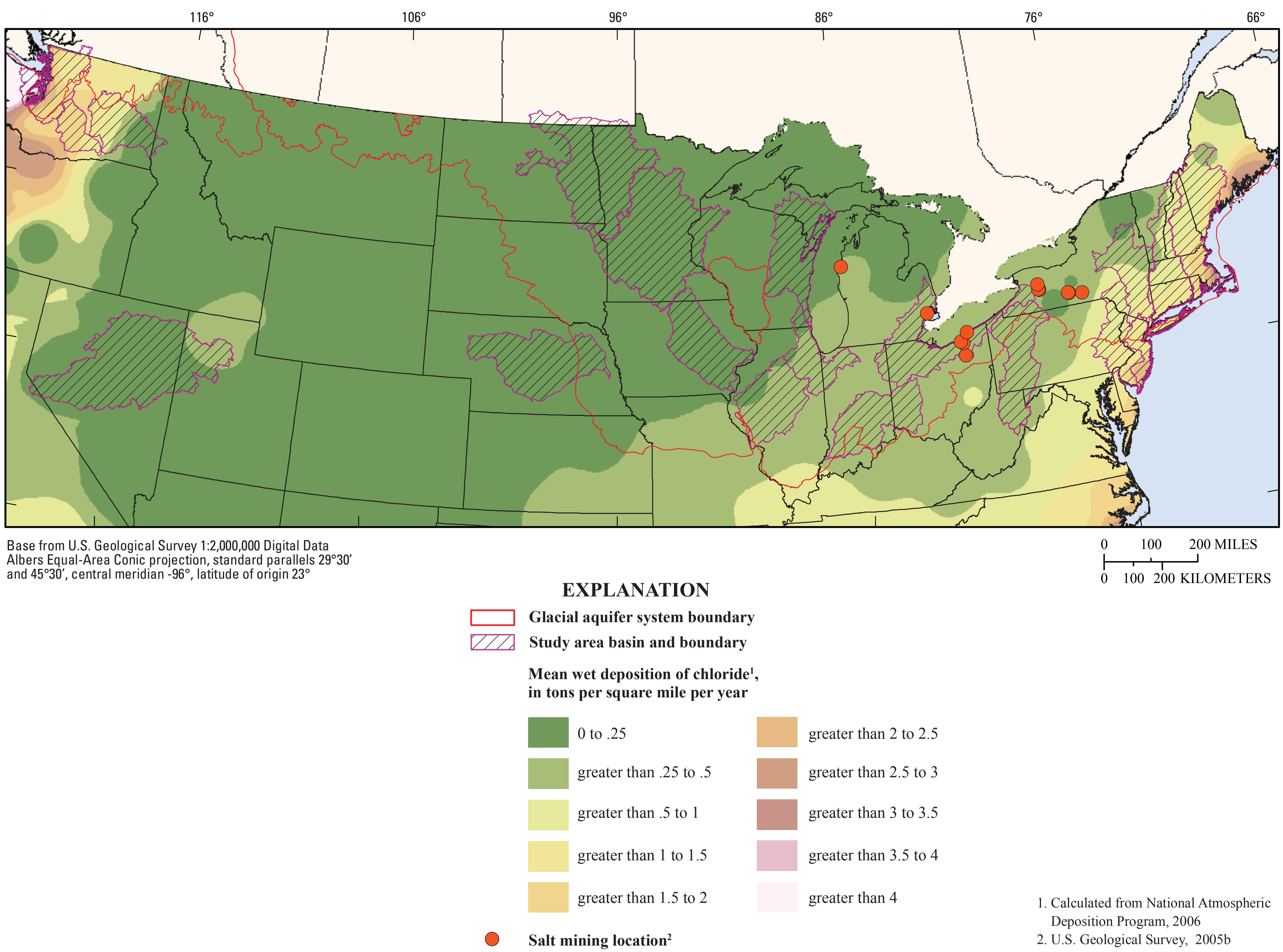


Figure 3. Mean wet deposition of chloride, 1994–2003, and locations of salt mining in the glacial aquifer system study area, northern United States.

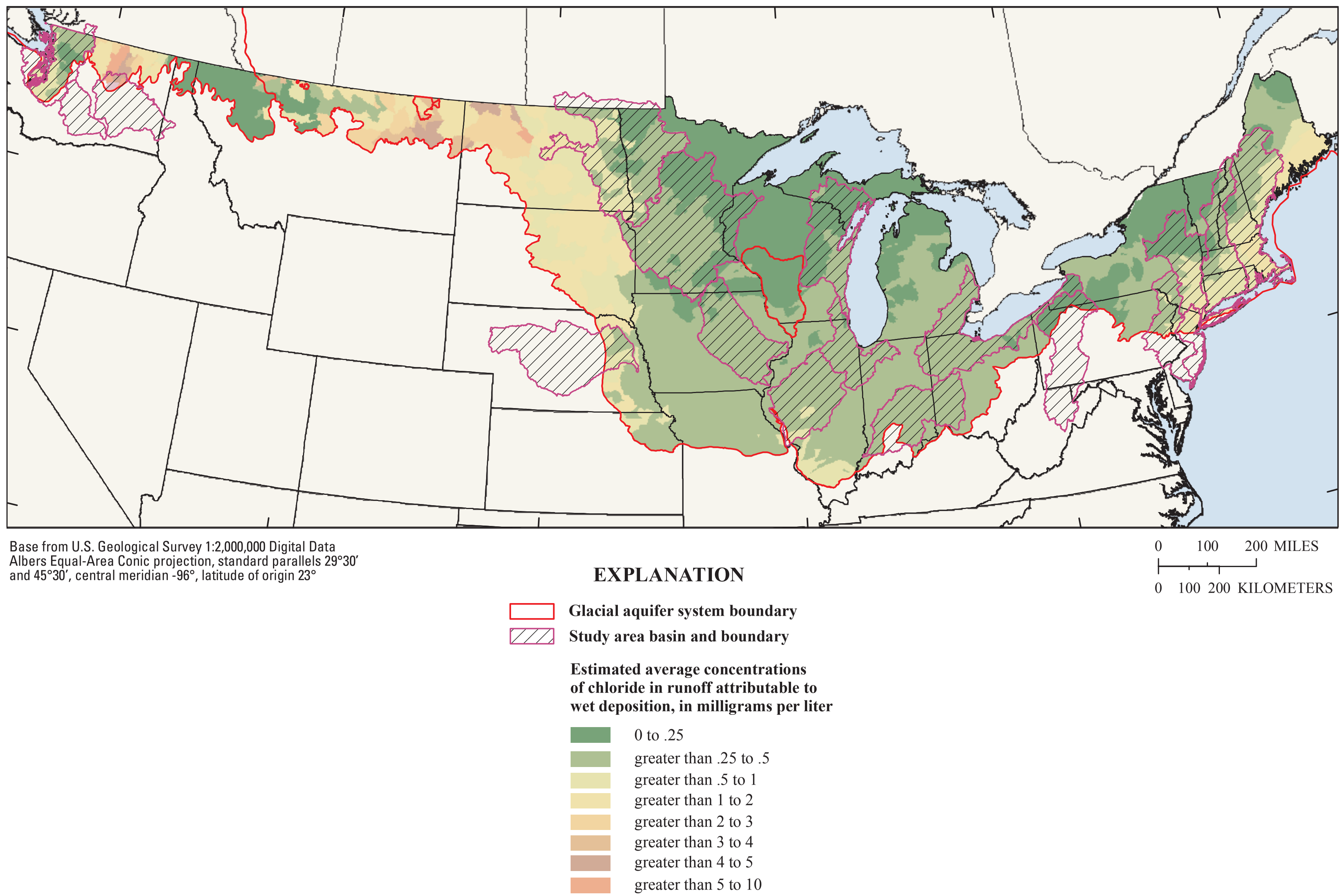


Figure 4. Estimated average concentration of chloride, 1994–2002, in runoff attributable to atmospheric (wet) deposition in the glacial aquifer system study area, northern United States. Data are combined from the National Atmospheric Deposition Program (2006) and runoff data from David Wolock, U.S. Geological Survey, written commun., 2006.

manufacturing (fig. 1B). The northern states that compose the glacial aquifer system (fig. 2) were the end destination for about 76 percent of the shipments of evaporated salt and rock salt (excluding brine) in the United States in 2005 (Kostick and others, 2007).

For many uses of salt, the chloride and sodium may end up being discharged to groundwater and surface waters as a direct or indirect result of use. The common pathways include atmospheric deposition; the dissolution of deicing salts from normal use on streets, parking lots, highways, and other paved surfaces; storage and handling of deicing salt; release of brines from salt, oil, and gas production; leaching from landfills; the treatment of drinking water and wastewater; and discharge of wastewater from treatment facilities or septic systems. The section below describes some of the common pathways for salt to enter the environment from human use.

Application of Deicing Salts

The use of deicing salts has improved the safety of winter weather driving, reducing accident rates by a factor of 8 on two-lane highways and 4.5 on multi-lane highways. The use of salt reduces injuries, property damage, and the severity of accidents during winter storms, thereby reducing costs of accidents (Kuemmel and Hanbali, 1992).

General highway application rates for deicing salt range from less than 1 ton per lane mile[2] per year to 20 tons per lane mile per year (Transportation Research Board, 1991; Jones and Sroka, 1997). Information on the rate of deicing-salt applications was acquired by compiling a list of the cities nearest to groundwater-sampling locations (table 2) and conducting an informal, qualitative survey of 20 local and State highway superintendents to provide information on the type and amount of deicing chemicals being applied to roads and highways in different parts of the study area. Respondents were asked to describe the number of lane or road miles serviced, and the type and amount of deicing chemicals purchased; application rates in units of tons per road or lane mile were calculated on the basis of that information (table 2).

Most of the highway superintendents reported the use of sodium chloride rock salt. Calcium and magnesium chloride were used in some cases as a wetting agent for rock salt. Application rates per lane mile ranged from less than 1 ton per road mile in Washington State to 74.5 tons per lane mile for a section of Interstate 84 in southeastern New York (Heisig, 2000). Most rates for deicing salt reported by highway officials ranged from 10 to 30 tons per lane mile (table 2). These values are similar to those reported in the literature.

In addition to the effects of application of deicing salts to impervious areas, salt-storage areas have been sites of historical contamination caused by runoff from uncovered salt piles, and infiltration when stored over pervious areas (Ostendorf and others, 2006). Many of these historical sites have been improved in recent years through the use of covered salt-storage buildings on impervious substrate. However, high chloride concentrations can persist in groundwater because of the long traveltimes from recharge areas to discharge at a well or surface-water body.

Highway maintenance crews in some parts of the study area are using techniques that enhance the effectiveness of deicing practices. These anti-icing techniques are designed to use salt more effectively by preventing a bond between ice and road surfaces rather than melting ice that has already bonded to the road. These updated practices include the use of road weather-information systems (RWIS) to decide which road deicing practices are appropriate based on weather and road temperature data and the use of salt that has been prewetted, or salt brine, on roadways prior to a predicted storm (Aultman-Hall and others, 2006).

Landfills

Salt from human consumption or activities is commonly deposited in municipal landfills. These salts typically are from food wastes and other products containing salt, including rubber, metals, and paper products. The estimated 10.3 million tons of salt or salt-bearing products deposited in landfills in 1990 represented 23 percent of reported consumption (Kostick, 1993). This estimated mass indicates that landfills can be source areas for salt constituents. Chloride concentrations are typically high in landfill leachate or in groundwater beneath or downgradient from landfills. For instance, the median chloride concentration in leachate from seven landfills in Illinois was 1,284 mg/L (Panno and others, 2006). The chloride concentration in leachate in a municipal landfill in Waterbury, Connecticut, was 724 mg/L in 1997 (Fuss & O'Neill, Inc., 1997), and 247 mg/L in shallow groundwater downgradient from the landfill (Mullaney and others, 1999). Landfills are commonly limited in aerial extent and can be areas with low groundwater recharge because of impervious capping material, which limits the release of large volumes of leachate to shallow aquifers or surface-water bodies. However, as with deicing-salt storage areas, landfills can be a long-term source of salt to aquifers and streams as a result of long groundwater traveltimes.

Wastewater and Water Treatment

Chloride in the environment is conservative (nonreactive), and thus there is little loss when chloride in salts contained in food, beverages, and household cleaning products is discharged to the environment through septic systems and wastewater-treatment facilities. In addition, water softeners used to treat water hardness commonly use salt brine to regenerate the resin in the treatment system with sodium, displacing calcium and magnesium. The remaining brine is then disposed of through the wastewater system, or to a dry well, where it may enter the underlying aquifer and ultimately discharge to a surface-water body.

[2] The lane mile is an areal measurement of road surface. The lane mile is one traffic lane (12 ft wide), extending for 1 mi along a road. When applied to highways, this measurement unit includes paved borders and breakdown lanes (Church and others, 1996).

Table 2. Selected annual application rates of deicing chemicals on State and local roads in the glacial aquifer system, northern United States.

State, municipality, or region	State	Road miles managed	Lane miles managed	Deicing chemical	Total amount of deicing salt used (tons)	Average use per lane mile (tons)	Average use per road mile (tons)	Source
Connecticut Department of Transportation	CT	3,276		Sodium chloride	101,947		31.1	Pat Rodgers, Connecticut DOT, oral commun., 2006
Manchester	CT	200		Sodium chloride	2,500		12.5	Kenneth Longo, Manchester Department of Public Works, oral commun., 2006
Woodbury	CT	86		Sodium chloride	900		10.5	Woodbury Department of Public Works, oral commun., 2006
Scituate Reservoir drainage basin–State Roads	RI	90	191	Sodium chloride	611	3.2	6.8	Nimiroski and Waldron, 2002, note that estimated sodium chloride reported based on calcium chloride/sodium chloride mixture
Scituate Reservoir drainage basin–Local Roads	RI	139	277	Sodium chloride	2,784	10.1	20.1	Nimiroski and Waldron, 2002
Iowa Department of Transportation–State Roads	IA			Sodium chloride		3.8		Transportation Research Board, 1991
Ames	IA	230		Sodium chloride	2,000		8.7	John Joiner, Director, Ames Iowa, Department of Public Works, oral commun., 2006
Cedar Rapids	IA		720	Sodium chloride	7,500	10.4		Department of Street Maintenance, oral commun., 2006
Waverly	IA	75		Sodium chloride	500		6.7	Brian Sullivan, Superintendent, Division of Streets, Public Works Department, Waverly, IA, oral commun., 2006
Illinois Department of Transportation	IL			Sodium chloride		6.6		Transportation Research Board, 1991
Elgin	IL	312		Sodium chloride, some calcium chloride liquid	6,000		19.2	City of Elgin, Illinois, Web page at *http://www.cityofelgin.org/index.asp?NID=180*
Gurnee	IL		210	Sodium chloride		14.3		City of Gurnee, Illinois, Web page at *http://www.gurnee.il.us/public_works/about.html#streetdivision*
Indiana Department of Transportation	IN			Sodium chloride	335,137			Indiana Department of Transporatation, 2004, Web page at *http://www.in.gov/dot/div/communications/2004annualreport/Safety.pdf*
Indiana Department of Transportation	IN			Sodium chloride		9.0		Transportation Research Board, 1991

Table 2. Selected annual application rates of deicing chemicals on State and local roads, in the glacial aquifer system, northern United States.—Continued

State, municipality, or region	State	Road miles managed	Lane miles managed	Deicing chemical	Total amount of deicing salt used (tons)	Average use per lane mile (tons)	Average use per road mile (tons)	Source
Columbus	IN			Sodium chloride	2,000		8.5	Steven Brown, City of Columbus, Indiana, Public Works, oral commun., 2006
Indianapolis Department of Transportation	IN	7,329	4,000	Cargill ClearLane (treated sodium chloride) liquid calcium chloride, magnesium chloride, and brine also used	48,000	12.0	6.5	John Burkhardt, City of Indianapolis, Department of Public Works, oral commun., 2006
State of Michigan Department of Transportation	MI			Sodium chloride		12.9		Transportation Research Board, 1991
Brighton	MI	25	50	Sodium chloride	1,505	21.0	42.0	Matthew J. Schindewolf, Director of Public Services, Brighton, Michigan, oral commun., 2006
Farmington Hills	MI	301		Sodium chloride	6,500		21.6	Daniel Rooney, Division of Public Works, Farmington Hills, Michigan, oral commun., 2006
Rochester Hills	MI		506	Sodium chloride	4,270	8.4	16.9	Roger Rousse, Department of Public Service, Rochester Hills, Michigan, oral commun., 2006
Wolverine Lake	MI	20		Sodium chloride	750		37.5	Andy Stone, Department of Public Serivce, Wolverine Lake, Michigan, oral commun., 2006
Minnesota Department of Transportation	MN	12,000		Sodium chloride	233,434 & 2.3 Mgal of brine		19.5	Minnesota Department of Transportation, 2006
Twin Cities Metro Area	MN		5,900	Sodium chloride	103,000	17.5		Minnesota Department of Transportation, 2006
Brooklyn Center	MN		139	Sodium chloride		27.0		Wenck Associates, Inc., 2004
Brooklyn Park	MN		243	Sodium chloride		27.0		Wenck Associates, Inc., 2004
Crystal	MN		112	Sodium chloride		3.0		Wenck Associates, Inc., 2004
Robbinsdale	MN		88	Sodium chloride		12.0		Wenck Associates, Inc., 2004

Table 2. Selected annual application rates of deicing chemicals on State and local roads, in the glacial aquifer system, northern United States.—Continued

State, municipality, or region	State	Road miles managed	Lane miles managed	Deicing chemical	Total amount of deicing salt used (tons)	Average use per lane mile (tons)	Average use per road mile (tons)	Source
New Hampshire Department of Transportation, Interstate 93	NH		111	Sodium chloride	2,762	24.9		Phil Trowbridge, New Hampshire Department of Environmental Services, written commun., 2006
New York, Croton Watershed–Interstate 84	NY			Sodium chloride		74.5	298.0	Heisig, 2000
New York, Croton Watershed–Taconic Parkway	NY			Sodium chloride		18.8	75.0	Heisig, 2000
New York, Croton Watershed Local Roads	NY			Sodium chloride		9.3	37.0	Heisig, 2000
Colonie Village	NY	35		Sodium chloride	1,500		43.5	Carl Fleshman, Superintendent of Public Works, Colonie Village, NY, oral commun., 2006
Ohio Department of Transportation	OH			Sodium chloride, calcium chloride also used		9.7		Kunze and Sroka, 2004 (for selected counties)
Harrison	OH		100	Sodium chloride, calcium chloride	600, 1,000 gal.	6.0		James Leslie, Director of Public Works, Harrison, OH, oral commun., 2006
West Carrollton	OH	300		Sodium chloride, calcium chloride	800		0.4	
Lacey City	WA		25	Sodium chloride	10	0.4		Dennis Ritter, Public Works Department, Lacey, WA, oral commun., 2006
Olympia	WA	200		Calcium magnesium acetate, some sodium chloride	5		0.03	Randy Stewart, Department of Transportation, Lacey, Washington, oral commun., 2006
Brookfield	WI		482	Sodium chloride	4,400	9.1	17.2	Terry Starns, Department of Public Works, Brookfield, Wisconsin, oral commun., 2006
Lake Geneva	WI		300	Sodium chloride	300	7.5		Lynn Allen, Department of Public Works, City of Lake Geneva, Wisconsin, oral commun., 2006
Milwaukee	WI		7,112	Sodium chloride	50,000	7.0		Hintz and others, 2001
Sussex	WI		45	Sodium chloride	1,500	33.3		Jeremy Smith, Department of Public Works, Sussex, Wisconsin, oral commun., 2006

A conservative estimate of the per capita salt consumption and chloride loss from wastewater can be made on the basis of the recommended daily adult sodium intake of 2,300 mg/d. The salt intake associated with 2,300 mg/d of sodium is 5,847 mg, which includes about 3,547 mg of chloride. This indicates that the average person on this diet would consume about 2.1 kg (4.7 lbs) of salt per year, and release about 1.3 kg (2.9 lbs) of chloride per year to wastewater discharge.

Water softeners can release considerably larger amounts of chloride to the environment. Discharge of chloride to the environment from water softening is through on-site septic systems, dry wells, or wastewater-treatment facilities. In 2005, salt for water treatment represented 3.1 percent of salt use in the United States (Kostick and others, 2007). Salt use varies in a typical residential water-softener installation, however, because of differences in water softeners, water use, and water hardness.

Water softeners likely are used extensively in the study area for homes with self-supplied domestic wells and in areas with publicly provided surface and groundwater supplies. Moderately hard water is found in most regions of the study area, although soft water predominates in the New England area. Hard and very hard waters are present in central and west central States, including Ohio, Michigan, Illinois, Wisconsin, Minnesota, and North Dakota (Briggs and Ficke, 1977).

Agriculture

Agricultural use of salt was about 3.5 percent of total use in 2005 (Kostick and others, 2007). Salt is used as an animal feed additive and may be used in other agricultural products such as pesticides and fertilizers. Salt from animal feeds may discharge to surface water and groundwater from feedlots and manure containment areas and from the use of manure as a fertilizer. Panno and others (2006) reported a median chloride concentration of 847 mg/L from hog and horse waste, and a median concentration of 57 mg/L in water samples from wells affected by animal waste in Illinois.

Agricultural fertilizers can be a source of chloride to receiving waters because the potassium in most fertilizers is in the form of potassium chloride. Other sources of chloride include the concentration and dissolution of salts resulting from irrigation from deep groundwater sources.

Methods of Data Analysis

NAWQA Program data from 1991 to 2004 from wells in the glacial aquifer system and from streams draining areas underlain by the glacial aquifer system were analyzed for this study. Ancillary information was synthesized from data compiled for each surface-water-quality station that was sampled in the NAWQA Program. Many surface-water monitoring stations had additional water-quality data collected by the USGS for State water-quality monitoring networks.

Data analysis included graphical plotting of concentrations of water-quality constituents, multiple-comparison tests, and statistical analyses of the relations between ancillary variables and chloride concentrations and yields in surface water. A multiple linear regression model (LOADEST) (Runkel and others, 2004) was used to analyze chloride loads in water from 95 surface-water monitoring stations.

Compilation of Environmental Data

Groundwater-quality and surface-water-quality data were synthesized from data collected from 1991 to 2004 in the study basins shown in figure 2. Groundwater-quality data were compiled from a national database established for all groundwater samples collected for the NAWQA Program in the glacial aquifer system study area. The groundwater data collected represent two different networks: (1) shallow monitoring wells in forested, agricultural, or urban land (land-use studies), and (2) drinking-water supply wells tapping glacial deposits (major aquifer studies) (Rosen and Lapham, 2008).

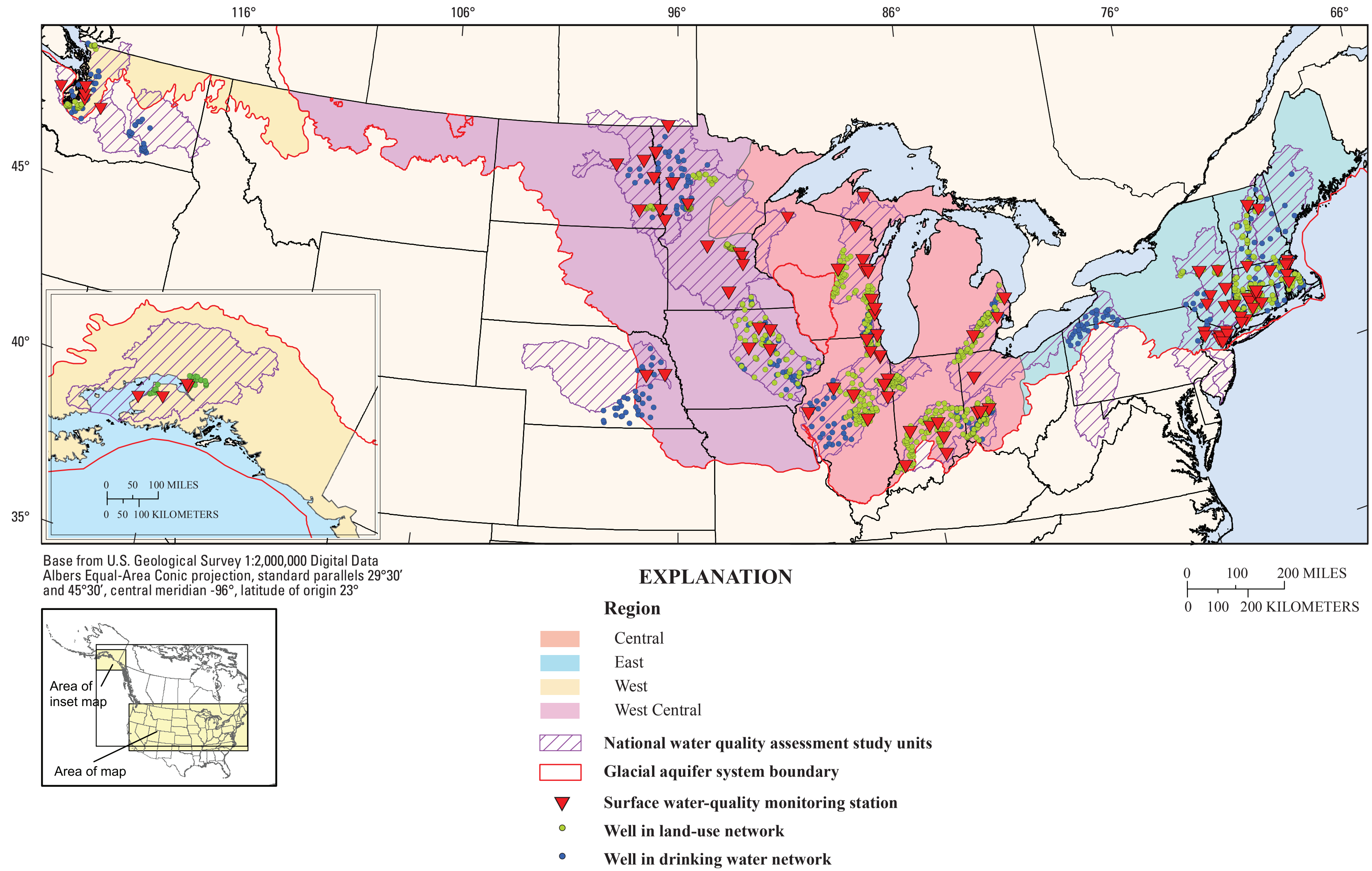
In land-use studies, data were collected from installed observation wells and selected existing observation wells to assess the quality of recently recharged shallow groundwater beneath different land-use settings (Gilliom and others, 1995). For this investigation, data from 797 wells from land-use studies (fig. 5) representing forested, agricultural, and urban areas were compiled for statistical analysis. Samples generally were collected only once from these wells.

In major aquifer studies conducted by the NAWQA Program, data were collected from 94 public and 438 private drinking-water supply wells in the glacial aquifer system (drinking-water network, fig. 5). These wells are typically deeper than the wells sampled for land-use studies and may integrate recharge from several land uses.

Water-quality data from 100 surface-water monitoring stations (Appendix 1) draining basins dominated by forested, agricultural, or urban land were selected for analysis (fig. 5). The contributing areas of these basins average 286 mi^2, and the median is 88 mi^2; of these basins, at least 31 contain major point-source wastewater discharges listed in the Permit Compliance System (PCS) database (U.S. Environmental Protection Agency, 2007). Surface-water monitoring stations had at least 20 samples collected over at least a 3-year period from 1991 to 2004. Samples generally were collected monthly for 3 to 13 years. Nineteen of the stations selected have long-term-flow and water-quality records that began in the 1960s to 1980s.

Multiple Comparison Testing

Multiple comparison tests were done by using S-PLUS 7 software (Insightful Corporation, 2005) to determine if populations of log base 10 chloride and sodium concentrations and chloride:bromide ratios by mass (Cl:Br) in groundwater were significantly different by adjacent land use or well type.



Base from U.S. Geological Survey 1:2,000,000 Digital Data
Albers Equal-Area Conic projection, standard parallels 29°30′ and 45°30′, central meridian -96°, latitude of origin 23°

Figure 5. Wells and surface-water-quality monitoring stations used in the analysis of the glacial aquifer system study area, northern United States.

This analysis also was used with maximum measured chloride concentrations and yields from samples collected in streams. A one-way analysis of variance (ANOVA) test was used to determine if the null hypothesis was rejected; the null hypothesis states that the means of all groups were not significantly different. If the null hypothesis was rejected, a Tukey's test for pairwise comparisons was done (Helsel and Hirsch, 2002).

Multiple Linear Regression Modeling

Multiple linear regression models were developed to describe maximum measured chloride concentrations (1991–2004) and mean chloride yield (1991–2003) for samples collected from surface-water monitoring stations. Explanatory variables that were evaluated for use in the models included land-use/land-cover characteristics, population density, road density, hydrologic and climatic variables, and wastewater discharges (table 3). Variables were selected on the basis of plausibility, statistical significance, and the distribution of residuals.

Data on land-use and land-cover characteristics were determined from LANDSAT Thematic Mapper (TM) images created for the National Land Cover Dataset (NLCD) (Vogelmann and others, 1998). The version of the NLCD (NLCDe) used was enhanced as described by Nakagaki and Wolock (2005).

Road density was computed as the length of all roads in a basin divided by basin area (Curtis Price, U.S. Geological Survey, written commun., 2006). Road density by category was compiled for surface-water basins from the digital Streetmap layer of the Tele Atlas North America (ESRI, Inc., 2006).

Potential evapotranspiration data for each surface-water site were developed through the use of the Parameter-elevation Regressions on Independent Slopes Model (PRISM) with the methodology described by Daly and others (1994).

Hydrologic variables were compiled from a national application of the TOPMODEL rainfall-runoff model (Wolock, 1993). Stream base-flow index was estimated by using data developed by Wolock (2003).

Estimation of Chloride Loads

Chloride loads in streams were estimated for 95 basins (those with sufficient data for load estimation)—21, 44, and 30 representing forested, agricultural, and urban as the dominant land use, respectively (Appendix 1). Loads were estimated for water years with available data from 1992–2003. Nineteen stations had data collected earlier than 1992, and chloride loads at these sites were plotted against time to identify obvious long-term trends.

Loads were estimated by using a multiple linear regression model included in the computer program LOADEST (Runkel and others, 2004). Given a time series of streamflow, additional data variables, and constituent concentration, LOADEST assists the user in developing a regression model for the estimation of constituent load. Explanatory variables in the regression model include various functions of streamflow, decimal time, and additional user-specified data variables. The formulated regression model then is used to estimate loads over a user-specified time interval. Mean load estimates, standard errors, and 95-percent confidence intervals are developed on an annual basis. The calibration and estimation procedures in LOADEST are based on three statistical estimation methods. The first two methods, Adjusted Maximum Likelihood Estimation (AMLE) and Maximum Likelihood Estimation (MLE), are appropriate when the calibration model errors (residuals) are normally distributed. Of the two, AMLE is the method of choice when the calibration data set (time series of streamflow, additional data variables, and concentration) contains censored data. The third method, Least Absolute Deviation (LAD), is an alternative to MLE when the residuals are not normally distributed (Runkel and others, 2004). Load estimates for selected urban basins were plotted to demonstrate visible trends with time by using a LOWESS smooth (Helsel and Hirsch, 2002).

Chloride in Groundwater and Surface Water

The concentrations of chloride and sodium in water from wells in the land-use studies network and major aquifer studies network were compared first by land use and well type and then to applicable drinking-water standards. Sources of chloride were evaluated on the basis of Cl:Br plotted against chloride and compared to binary mixing curves estimated for different sources.

Surface-water-quality data were compared with the recommended criteria for chloride concentrations to protect aquatic life and to determine the months when the recommended criteria concentrations commonly were exceeded. A multiple linear regression model was developed to describe the variability of the natural log of the maximum measured chloride concentration at all surface-water monitoring stations. These maximum measured concentrations of chloride were compared with concentrations of chloride in base-flow samples from each station to determine if base-flow concentrations could be used to predict which stations would have chloride concentrations greater than recommended aquatic criteria. Lastly, the loads and yields of chloride from 95 monitoring stations were computed, and yields were compared by dominant land use. A multiple linear regression model was used to describe the variability of the natural log of chloride yield. Selected stations with long-term data were used to show examples of apparent trends in chloride loads with time.

Table 3. Watershed characteristics evaluated in a regression analysis of maximum measured chloride concentrations.

[NLCDe, National Land Cover Dataset, Vogelmann and others, 1998, enhanced as described by Nakagaki and Wolock (2005); km, kilometer; km^2, square kilometer; cm, centimeter; mi, mile; mi^2, square mile; PRISM, Parameter-elevation Regressions on Independent Slopes Model; PCS, permit compliance system]

Land use, land cover (Vogelmann and others, 1998), population and road density
Percent of basin composed of low intensity residential, NLCDe
Percent basin composed of high intensity residential, NLCDe
Percent basin composed of commercial/industrial/transportation, NLCDe
Percent of basin composed of orchards/vineyards/other, NLCDe
Percent basin composed of pasture/hay, NLCDe
Percent basin composed of row crops, NLCDe
Percent basin composed of small grains, NLCDe
Percent basin composed of fallow, NLCDe
Population density 2000, in people/km^2 (ESRI, 2006)
Road density in km/km^2 (Curtis Price, U.S. Geological Survey, written commun., 2006)
Road density by type (ESRI, Inc., 2006)
Major highway density, in mi/mi^2
Major highway connector density, in mi/mi^2
Highway density, in mi/mi^2
Major road density, in mi/mi^2
Local road density, in mi/mi^2
Hydrologic and climatic variables
Percent of basin streamflow contributed by Dunne overland flow (estimated by means of TOPMODEL (Wolock, 1993) hydrologic model)
Percent of basin streamflow contributed by Horton overland flow (estimated by means of TOPMODEL (Wolock, 1993) hydrologic model)
Mean subsurface contact time in days (estimated by means of TOPMODEL (Wolock, 1993) hydrologic model)
Base-flow index
Mean potential evapotranspiration in cm (PRISM) [Daly and others, 1994]
Wastewater discharges
Number of major discharges upstream of monitoring site, PCS database (U.S. Environmental Protection Agency, 2007)
Number of minor discharges upstream of monitoring site, PCS database (U.S. Environmental Protection Agency, 2007)

Groundwater

Groundwater samples collected from wells in the land-use studies contained different distributions of chloride depending on the dominant land use (fig. 6). The largest median concentration of chloride was in samples from urban land-use wells (46 mg/L) and was about 16 times larger than the median concentration in samples from forested land-use wells (2.9 mg/L). The median concentration in water samples from agricultural land-use wells (12 mg/L) was about four times larger than in samples from forested land-use wells. The median concentration of chloride in drinking-water supply wells was 26 mg/L in public-supply wells and 12 mg/L in private domestic wells (fig. 6). The results of a one-way ANOVA indicated that the means of the populations of log base-10 transformed chloride data were significantly different. A Tukey's test indicated that only the agricultural land-use wells and the private drinking-water wells were not significantly different (fig. 6).

Chloride concentrations in groundwater were greater than the SMCL of 250 mg/L for drinking water (U.S. Environmental Protection Agency, 1992) in samples from 20 of 797 land-use wells (2.5 percent); 17 were urban land-use wells and 3 were agricultural land-use wells. Chloride concentrations were greater than 250 mg/L in 10 of 532 drinking-water wells (1.7 percent); 9 were private domestic wells and 1 was a public-supply well. Groundwater with concentrations of chloride in excess of 230 mg/L that discharges to surface water may cause toxic effects to aquatic life on the basis of the recommended criteria established by U.S. Environmental Protection Agency (1988).

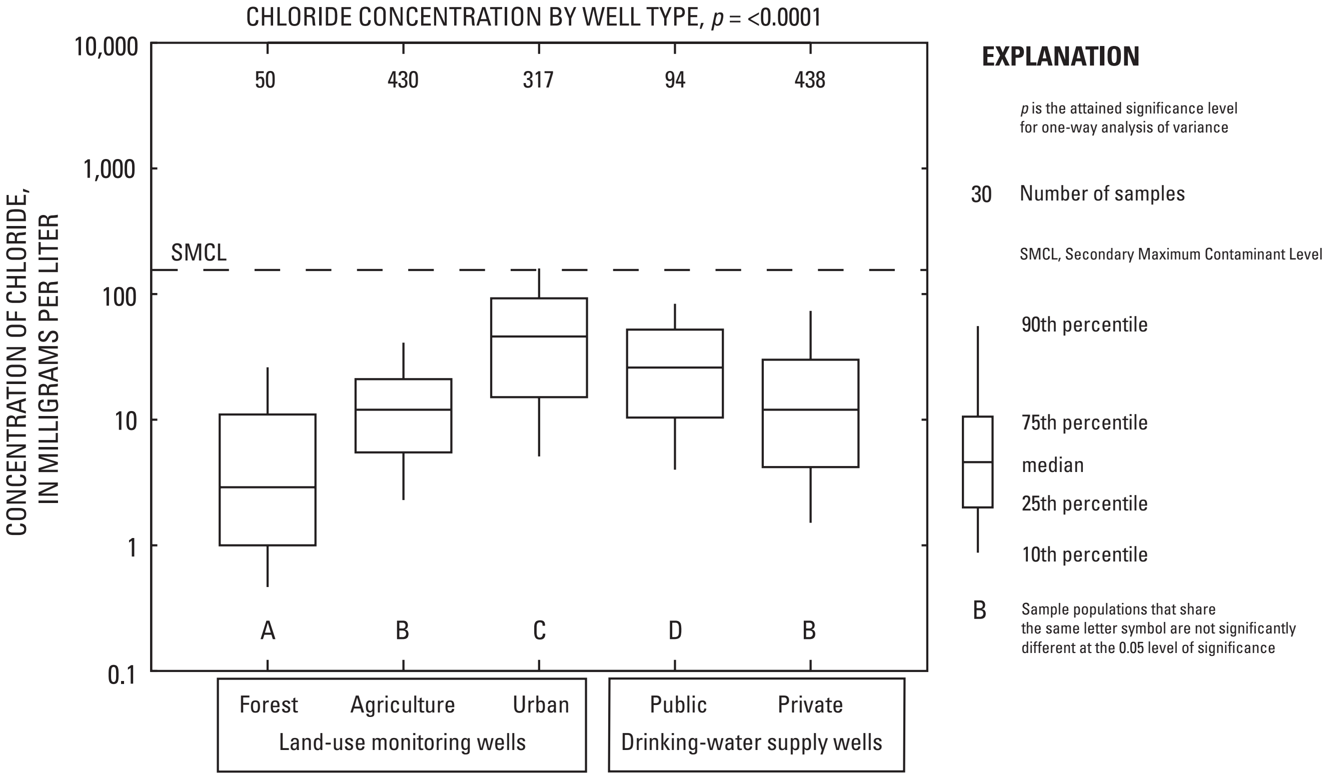


Figure 6. Distribution of chloride concentrations in samples from shallow monitoring wells in forested, agricultural, and urban areas and in drinking-water supply wells in the glacial aquifer system, northern United States, 1991–2003.

Because sodium in drinking water is considered a human health issue, concentrations of sodium were also summarized for groundwater sites in this study. The distribution of sodium concentrations (in samples from 1,332 sites) was similar to that of chloride in the land-use wells. The largest median concentration of sodium was in samples from urban-land-use wells (26 mg/L) and was more than six times larger than the median concentration of sodium (4.1 mg/L) in water samples from forested-land-use wells. The median sodium concentration in water samples from agricultural-land-use wells (6.3 mg/L) was larger than the median sodium concentrations in the forested-land-use wells (fig. 7). The median sodium concentration was 19 mg/L in public-supply wells and 12 mg/L in private domestic wells. The only Federal drinking-water standard for sodium is an unenforceable Drinking Water Advisory of 20 mg/L for individuals on a 500 milligram per day (mg/d), low-sodium diet (U.S. Environmental Protection Agency, 2004). Water in the majority of urban land-use wells (57.1 percent) had concentrations of sodium equal to or greater than 20 mg/L, followed by agricultural land-use wells (16.7 percent) and forested land-use wells (8.0 percent). The concentration of sodium was equal to or larger than 20 mg/L in samples from 46.8 percent of the public-supply wells and 33.9 percent of the domestic wells.

Sources of Chloride in Groundwater

Chloride:bromide ratios (Cl:Br) have been used by several researchers to identify the sources of chloride in groundwater (Davis and others, 1998; Thomas, 2000; Jagucki and Darner, 2001; Panno and others 2006). The method is based on understanding the Cl:Br and chloride concentrations of different chloride sources, and the resulting Cl:Br and chloride concentrations when these sources are mixed.

Groundwater samples collected from a network of forested land-use wells had the lowest median Cl:Br ratio (by mass) (148). Samples collected from wells in the urban land-use study had the highest median Cl:Br (879), followed by the samples from wells in the agricultural land-use well study (283) (fig. 8). The median Cl:Br of water samples from the private drinking-water wells (193) was not significantly different from the median for forested land-use wells, and the median Cl:Br of samples from the public drinking-water-supply wells (673) was not significantly different from the ratio for the urban land-use wells (fig. 8).

Mixing curves (fig. 9A) were developed to represent binary mixtures of halite, sewage or animal waste, potassium chloride fertilizers, landfill leachate, basin brines, and seawater sources of chloride on the basis of data and studies listed in table 4. Binary mixing-curve lines were determined by using

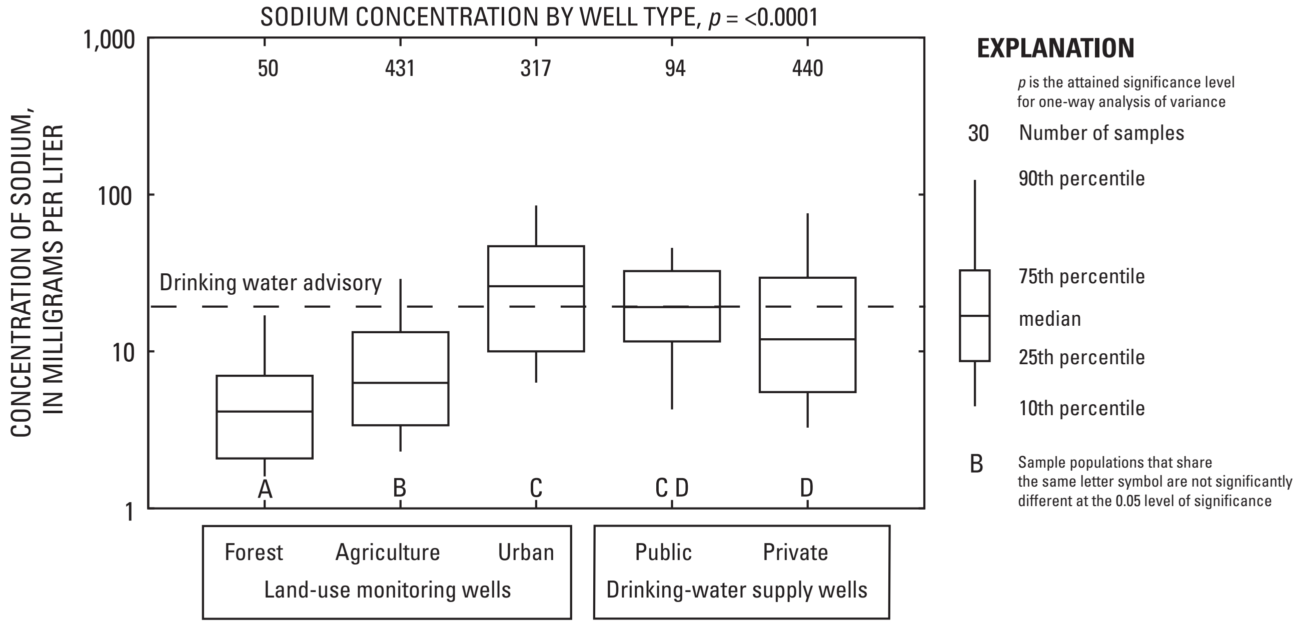


Figure 7. Distribution of sodium concentrations in samples from shallow monitoring wells in forested, agricultural, and urban areas and in drinking-water supply wells in the glacial aquifer system, northern United States, 1991–2003.

mixing equations described by Jagucki and Darner (2001). Data from the land-use wells were plotted with the binary-mixing curves to evaluate if any of samples were representative of end members or simple mixtures (figs. 9B–D).

Points representing groundwater samples that do not fall on any of the binary mixing lines reflect the reality that there may be more than two sources of chloride, and the actual mixtures in groundwater samples are not likely mixtures of full-strength end members, but have been diluted with additional water. The effect of dilution of a concentrated end member with precipitation or dilute groundwater will be to retain the Cl:Br, but reduce the chloride concentration. An example of this would be a mixture containing chloride from sewage/animal waste and halite. The binary mixing curves 5 and 6 on figures 9A–D are based on the assumption that mixtures can contain different proportions of the full-strength end members. The halite end members were assumed to have a chloride concentration of 10,000–20,000 mg/L; higher concentrations are possible based on the solubility of halite. In reality, deicing salt is likely to be diluted with precipitation before recharging the groundwater, and septic-system waste may also be diluted with precipitation or dilute groundwater before moving downgradient. Therefore, most mixtures are not likely equivalent to binary mixtures, unless samples are collected very close to the source of the chloride.

The samples from the forested-land-use wells generally had low Cl:Br and low concentrations of chloride; however, the positions of the plotted points for nine samples (fig. 9B) indicated human influence from halite or sewage and animal waste. Samples from the agricultural-land-use wells were not dominated by any one source of chloride (fig. 9C), although there was a large concentration of points around mixtures of dilute groundwater and potassium chloride or sewage and animal waste. Sources of chloride in agricultural areas could include deicing salt; salt used in animal feeds, fertilizer, and water softeners; salt in animal waste; and the use of deep groundwater (containing brines) for irrigation. Points for chloride concentrations above 100 mg/L in samples from agricultural-land-use wells plotted near the binary mixing curves (lines) for dilute groundwater and (1) halite, or (2) sewage or animal waste mixed with halite, or (3) dilute groundwater mixed with basin brines, or (4) landfill leachate.

Points representing samples from the urban-land-use wells plotted along or between the mixing lines for dilute groundwater/halite, or dilute groundwater/sewage or animal waste, and dilute groundwater and potassium chloride fertilizer (fig. 9D). Many samples had a Cl:Br of 1,000 or higher and chloride concentrations greater than 100 mg/L. This analysis shows that, as in the agricultural areas, there are mixed sources of anthropogenic inputs of chloride to groundwater. Samples with a Cl:Br greater than 1,000 and chloride concentrations greater than 100 mg/L are likely to be dominated by halite inputs from deicing or water softeners.

Points representing samples from drinking-water supply wells initially were plotted with the binary mixing curves that were used in figures 9A–D; however, there was a wide

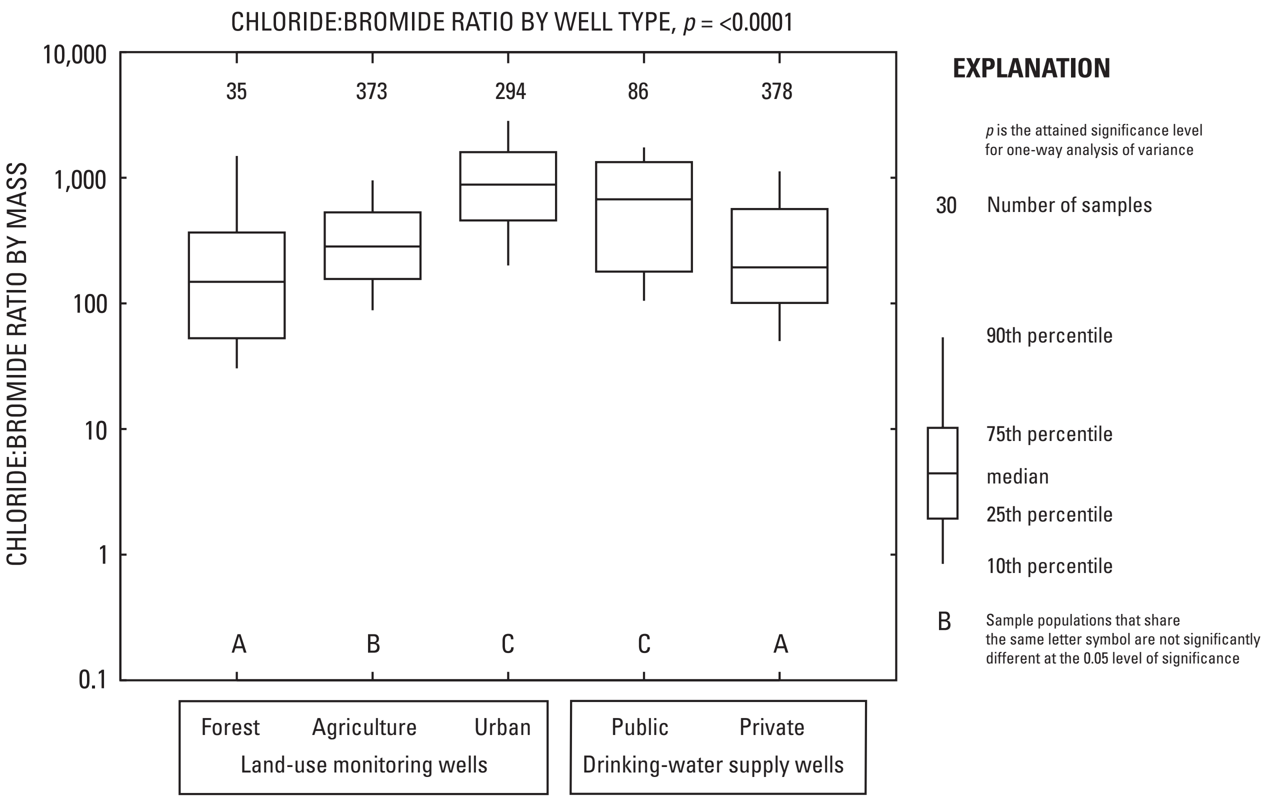


Figure 8. Distribution of chloride:bromide ratios (by mass) in samples from shallow monitoring wells in forested, agricultural, and urban areas and in drinking-water supply wells in the glacial aquifer system, northern United States, 1991–2003.

scattering of points around the binary mixing lines. There were, however, some statistically significant differences in the Cl:Br by supply-well type (fig. 8) and by the geographic region (fig. 10). Public drinking-water supply wells had a significantly higher Cl:Br than private drinking-water wells, indicating that water quality in these public drinking-water supply wells is influenced by human activities, primarily deicing, water softening, and sewage/animal waste.

The Cl:Br in samples from the private drinking-water supply wells was were not significantly different from ratios in samples from the forested land-use wells (fig. 8); however, concentrations of chloride were significantly higher in the private drinking-water supply wells than in samples from the forested land-use wells (fig. 6).

Samples from drinking-water supply wells in the east had significantly higher Cl:Br than samples from other regions in the glacial aquifer system (fig. 10). Samples from the west-central and west parts of the glacial aquifer system had the lowest Cl:Br (fig. 10). The Cl:Br in 65 percent of the samples from the east and 43 percent of the samples from the central parts of the glacial aquifer system were greater than 460 (table 4, minimum value for sewage and animal waste), indicating the influence of chloride from deicing salt, water-softening salt, and sewage and animal waste (fig. 10).

Surface Water

Surface-water-quality data from monitoring sites on 100 streams or rivers that were sampled 10 or more times for chloride from 1991 to 2004 were analyzed to make comparisons of chloride concentration to basinwide land use. Samples generally were collected monthly for 3 to 13 years. Some sites that were part of State networks had many more years of chloride data. Multiple linear regression models were developed to describe the distribution of maximum measured chloride concentrations and chloride yields for samples collected from 1991 to 2004 in forested, agricultural, and urban basins. Chloride yields were compared among forested, agricultural, and urban basins.

To evaluate the potential for chloride concentrations to be greater than the USEPA recommended aquatic life criteria described below for chloride (U.S. Environmental Protection Agency, 1988), the maximum measured concentrations (samples collected from 1991 to 2004) were compared by land use (fig. 11). The recommended chronic criterion for aquatic life is a 4-day average of 230 mg/L with a recurrence interval of once every 3 years (U.S. Environmental Protection Agency, 1988). Samples from 13 sites with urban land use and 2 sites with agricultural land use had chloride values that were greater than the chronic concentration value at least once (table 5). Sampling frequency was insufficient to determine whether the 4-day average would have been greater than 230 mg/L with a recurrence interval of less than once every 3 years. Six of the sites had chloride concentrations greater than 230 mg/L in 10 percent or more of the samples collected. Concentrations in samples from three sites were greater than the recommended acute aquatic criterion concentration for chloride of 860 mg/L (U.S. Environmental Protection Agency, 1988). This criterion relates to a 1-hour average concentration with a recurrence interval of less than once every 3 years.

Samples with concentrations of chloride greater than 230 mg/L generally were collected during the winter and spring months, primarily November–April, indicating a possible relation with winter deicing activity (table 5). Concentrations above the recommended criteria may occur during a rain or freezing rain event following the application of deicing chemicals, or when daytime temperatures rise high enough to melt roadside and parking-lot snow and ice (fig. 12). Samples with chloride concentrations above the recommended aquatic criteria in late spring and summer may be explained by the discharge of groundwater containing high concentrations of chloride (for example, Shingle Creek, station 05288705 and Lincoln Creek, station 040869415; table 5) or could be related to wastewater discharges with high chloride during a low-flow period, as in the case of two monitoring stations in Illinois (05531500, 05532000) identified in table 5 (CH2M HILL Inc., 2003).

Concentrations of chloride considered to represent base-flow samples collected from 1991–2004 were selected from the data by using the sample with the lowest river discharge at each site. Samples collected during base-flow periods are presumed to represent the quality of groundwater, except for basins with major wastewater discharges. The median concentration of chloride was 81 mg/L in base-flow samples from urban basins, 21 mg/L from agricultural basins, and 3.5 mg/L from forested basins. Chloride concentrations for the selected base-flow samples were plotted against the maximum measured chloride concentrations to determine whether there was a relation (fig. 13). Maximum measured chloride concentrations were greater than 230 mg/L in samples from sites at which base-flow chloride concentrations were greater than about 75–90 mg/L. This result suggests that base-flow concentrations can be used to predict which streams may have concentrations larger than the USEPA recommended chronic criteria for chloride. Streams with elevated concentrations of chloride in base flow may be more likely to exceed the recommended criteria during the winter months, because the chloride concentrations are high at the beginning of snowmelt events that contribute stormwater discharge containing concentrated chloride. Therefore, the loads in base flow from groundwater discharge and wastewater discharge are an important consideration in basins with chloride concentrations larger than 230 mg/L.

Relation of Maximum Chloride Concentration to Explanatory Variables

A multiple linear regression model was developed to describe the variability of the natural log of maximum measured chloride concentrations. The regression analysis was performed on chloride data from 83 of the 100 stations previously described. This subset was used because data for some of the explanatory variables (table 3) were unavailable for some stations. The data set included 15 forested basins, 41 agricultural basins, and 27 urban basins. Three significant variables—major road density, potential evapotranspiration, and the percentage of the annual streamflow from saturated overland flow (Dunne overland flow), (table 6)—explained 66 percent of the variability in the natural log of the maximum measured chloride concentration. Model residuals were approximately normally distributed with a constant variance (fig. 14).

Basins with greater density of major roads had higher maximum chloride concentrations. This variable indicates a likely relation with areas receiving deicing salts, but road densities also are usually correlated with overall urbanization and population density, indicating the possibility for multiple sources of chloride, as described in earlier sections of this report.

Basins with higher average potential evapotranspiration generally had higher maximum concentrations of chloride. The significance of this variable may be explained as follows: sites with low evapotranspiration are more likely to have more groundwater recharge (Granato and others, 1995), and areas with high potential evapotranspiration may be more likely to concentrate salts in basin recharge and runoff.

The variable “Dunne overland flow” was significant and had a negative sign. This variable is also known as “saturation overland flow” and was described by Dunne and Black (1970). The negative sign indicates that basins with the most annual saturated overland flow had lower maximum chloride concentrations, because with more overland runoff, there is a greater potential for dilution of salts.

Loads and Yields of Chloride from Forested, Agricultural, and Urban Basins

Chloride loads were estimated for 20 forested, 44 agricultural, and 31 urban basins from 1991 to 2003. The average load was determined from the LOADEST results for the number of years of available data within the study time period. Loads were normalized by drainage area and converted to yields in tons of chloride per square mile. The median yield was 6.4 tons/mi^2 from the forested basins, 15.4 tons/mi^2 from the agricultural basins, and 88 tons/mi^2 from the urban basins (fig. 15).

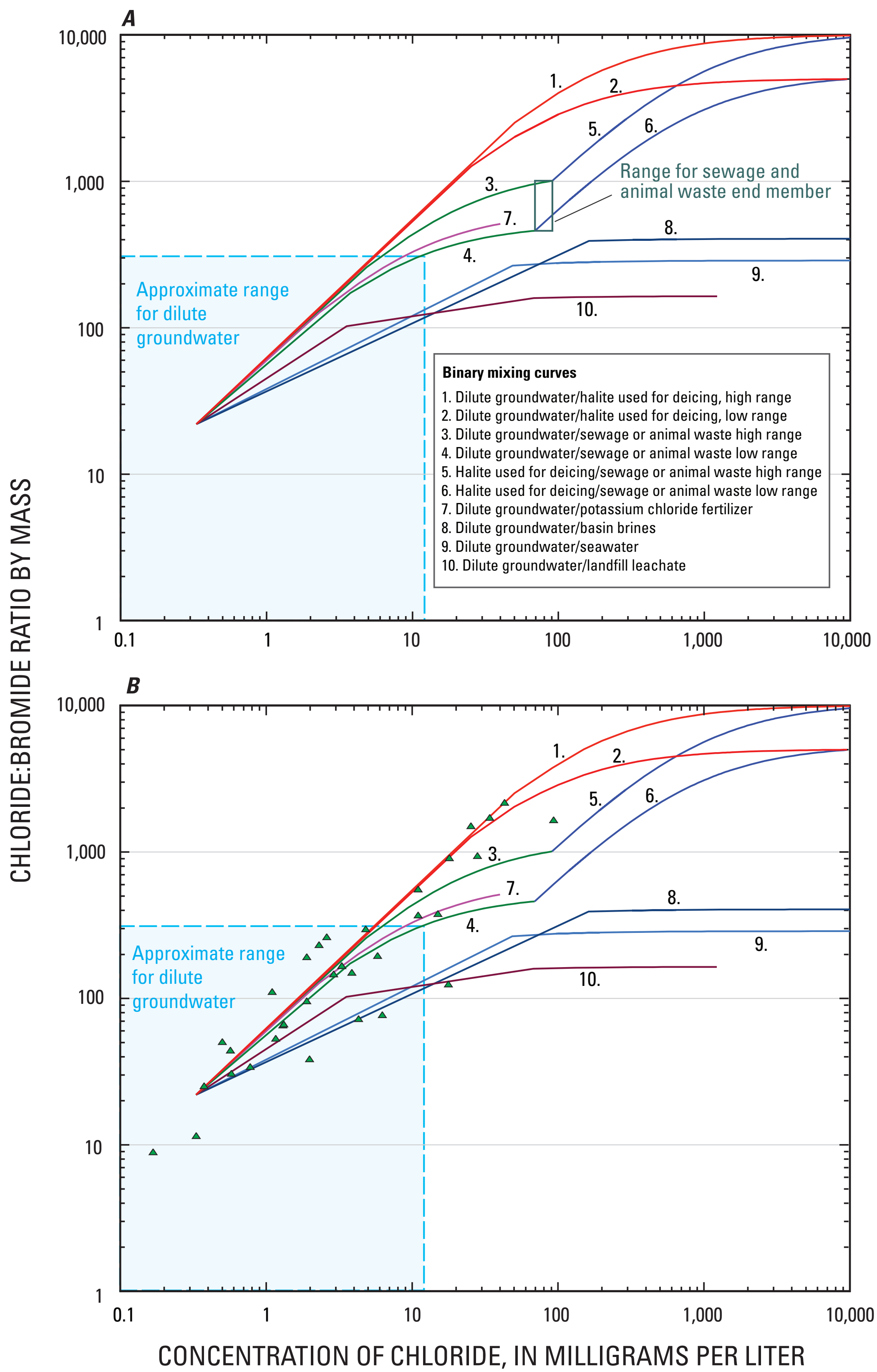


Figure 9. *(A)* Binary mixing curves representing sources of chloride. The relation of chloride concentration to chloride:bromide ratios (by mass) for samples from shallow monitoring wells in *(B)* forested areas, *(C)* agricultural areas, and *(D)* urban areas.

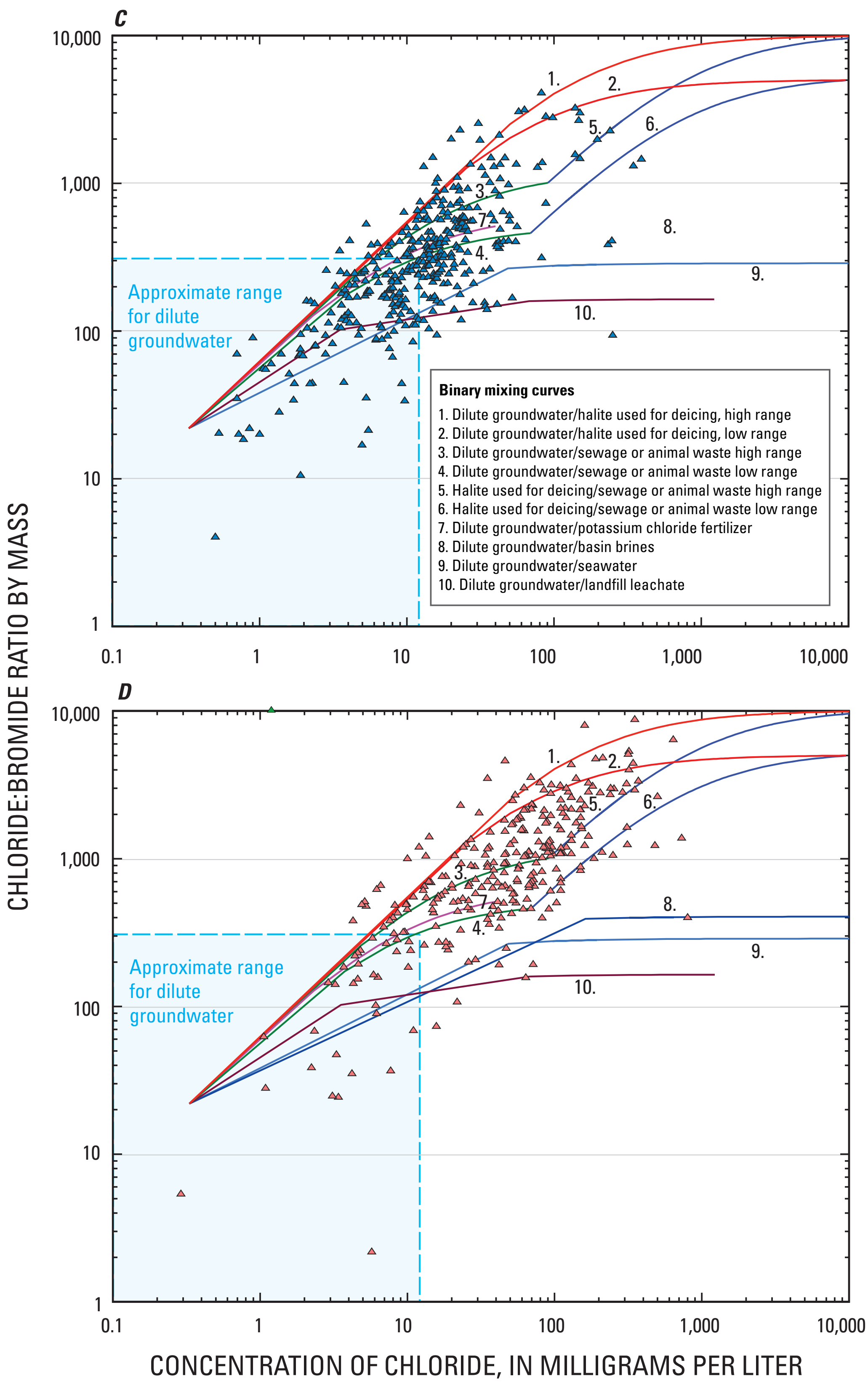


Figure 9. *(A)* Binary mixing curves representing sources of chloride. The relation of chloride concentration to chloride:bromide ratios (by mass) for samples from shallow monitoring wells in *(B)* forested areas, *(C)* agricultural areas, and *(D)* urban areas.—Continued

Table 4. Chloride:bromide ratios for sources of chloride to water resources.

[mg/L, milligrams per liter; Cl:Br, chloride to bromide ratio by mass; shaded areas represent end members used to draw binary mixing curves]

Chloride source and reference	Chloride (mg/L)	Bromide (mg/L)	Cl:Br
Dilute groundwater			
Forested land-use wells, glacial aquifer system (median of 35 samples)	3	0.02	148
End member used for dilute groundwater, from forested land-use well network	.33	.015	22
Sewage and animal waste			
Panno and others, 2006, median of samples from 6 private septic systems	91	.09	769
			[1]1,011
Davis and others, 1998 (assembled for U.S., England)	--	--	300–600
Thomas, 2000; Jagucki and Darner, 2001	69	--	460
Peavy, 1978	37–101	--	--
Panno and others, 2006, median of 4 samples of hog and horse waste	847	.570	1,395
Halite			
Granato, 1996; analysis of deicing salt	--	--	5,000
End member range used in binary mixing curves for deicing salt	[2]10,000–20,000		
Knuth and others, 1990, analysis of deicing salt	--	--	8,400
Davis and others, 1998	--	--	1,000–10,000
Panno and others, 2006, analysis of deicing salt	--	--	13,497
Panno and others, 2006, analysis of water softening salt	--	--	5,139
Panno and others, 2006, analysis of water softening salt	--	--	3,438
Seawater			
Feth, 1981	19,300	67	288
Landfill leachate			
Panno and others, 2006, median of 10 leachate samples from Illinois	1,284	7.8	193
			[3]164
Mullaney and others, 1999, landfill leachate affected groundwater sample	247	1.4	173
Fertilizer			
Panno and others, 2006, potassium chloride fertilizer sample analysis	[4]40	--	510
Basin brines			
Panno and others, 2006, Illinois Basin brines summarized from other studies	64,600	--	406

[1] Cl:Br ratio based on dividing Cl and Br values reported by Panno and others (2006).

[2] Assumed range in concentrated deicing salt runoff in binary mixing curves used.

[3] Cl:Br ratio based on dividing Cl and Br values reported by Panno and others (2006).

[4] Estimated upper limit based on analysis in Panno and others (2006).

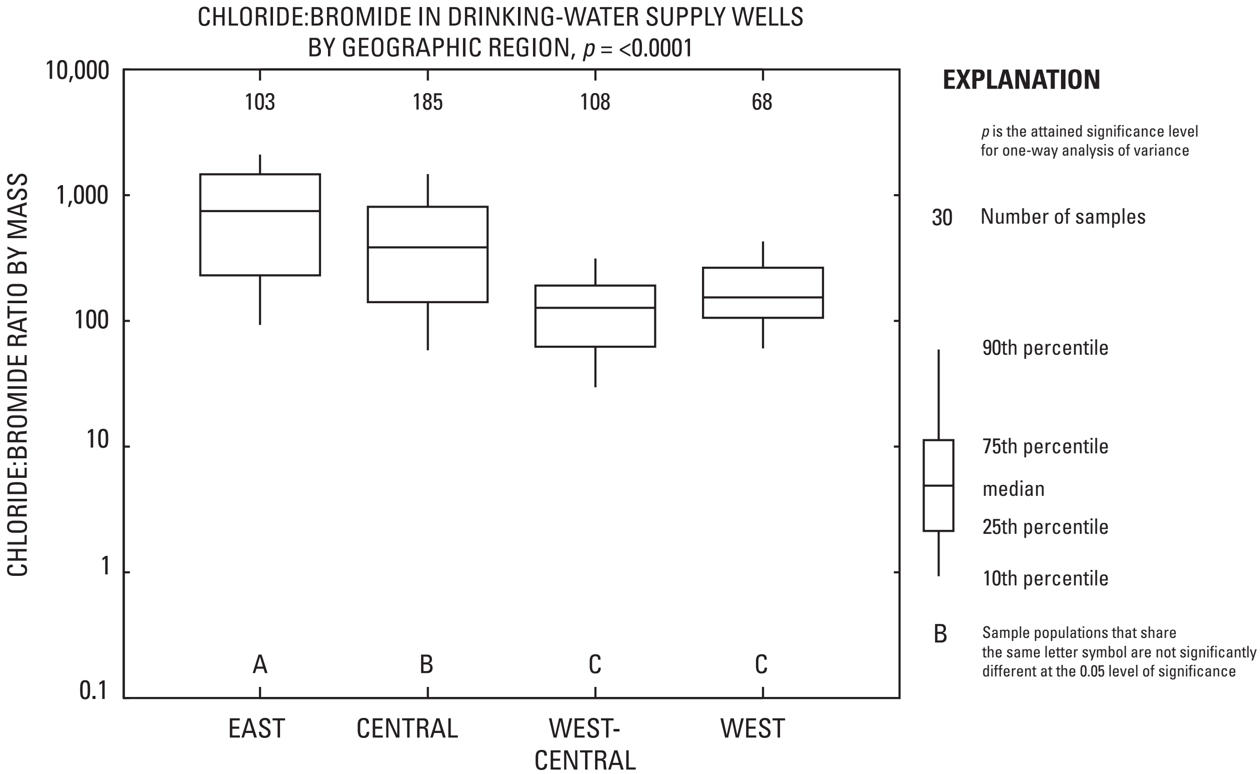


Figure 10. Distribution of chloride:bromide ratios (by mass) in samples from drinking-water supply wells in geographic regions of the glacial aquifer system, northern United States, 1991–2003.

Relation of Chloride Yield to Explanatory Variables

A multiple linear regression model was developed to describe the natural log of chloride yield. The data set included 15 forested basins, 41 agricultural basins, and 27 urban basins. Four variables (table 7) explained 69 percent of the variability in the natural log of chloride yield. Significant variables selected or modified from table 3 and shown in table 7 were Highway density and the Number of major discharges upstream of monitoring site in USEPA PCS database + (plus) 1, potential evapotranspiration, and the difference between the percent of urban and agricultural land. Model residuals were approximately normally distributed, with a constant variance (fig. 16).

Major dischargers include municipal wastewater-treatment facilities with discharges greater than 1 million gallons per day (Mgal/d) and other facilities with discharges that have been rated by USEPA as major, based on volume and type of pollutants and type of receiving waters (Steven Winnett, U.S. Environmental Protection Agency, written commun., 2007). The variable "number of major discharges upstream of the monitoring site in the USEPA Permit Compliance System (PCS) database," if used alone in a simple linear regression model, explains about 17 percent of the variability in the chloride yield, indicating that wastewater discharges are a likely component of chloride loads in some streams sampled.

The significance of the variable Highway density (table 3) is probably related to areas that receive deicing salts in winter. The density of different road categories is generally correlated with urban land use and population density, and therefore other sources of chloride from urban settings cannot be excluded from this analysis. The difference between percent urban and percent agricultural land was only weakly correlated (correlation coefficient 0.44) with highway density, and explained an additional 14 percent of the variability in chloride yield.

Trends in Loads at Selected Sites and Evaluation of Sources of Chloride

Long-term data were available for 19 of the monitoring sites for which loads were estimated. Load estimates for three selected urban basins with long-term data were plotted to demonstrate visible trends with time by using a LOWESS smooth (Helsel and Hirsch, 2002) (figs. 17A–C). The apparent trends are similar to those reported by Kaushal and others (2005). Increases in chloride load over time generally can be

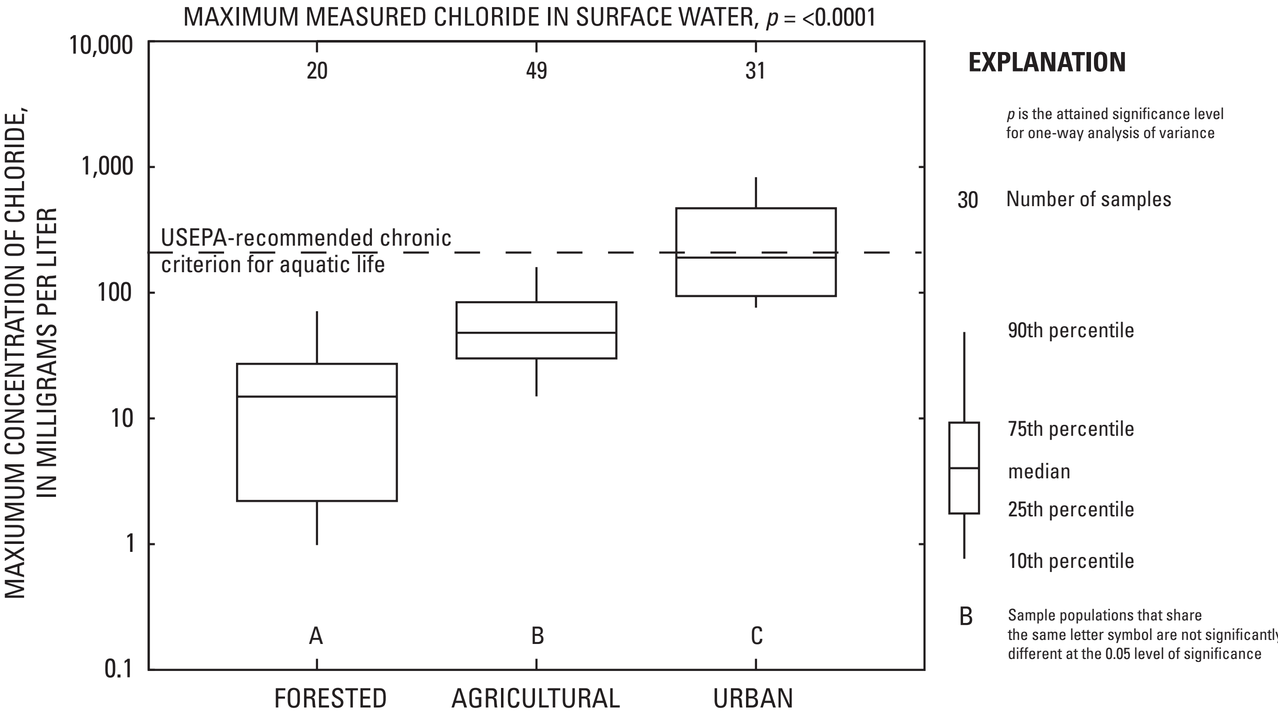


Figure 11. Distribution of maximum chloride concentrations measured at selected surface-water-quality monitoring stations in the glacial aquifer system, northern United States, 1991–2004.

attributed to changes in the application of deicing salt, the expansion of road networks and impervious areas that require deicing, increases in the number of septic systems, increases in the volume of wastewater discharge, and the arrival of saline groundwater plumes from landfills and salt-storage areas over time. These trends suggest that chloride concentrations in some urban basins may be greater than USEPA-recommended criteria in the coming years and decades.

Of the examples shown in figure 17, only Poplar Creek at Elgin, Illinois, receives no major wastewater discharges (Sullivan, 2000). The Rahway River near Springfield, New Jersey, may have some combined sewer overflows (J.G. Kennen, U.S. Geological Survey, written commun., 2006) and has 29 minor discharges listed in the PCS database.

The Quinnipiac River Basin in Connecticut was studied in greater detail to evaluate changes that have taken place during the period of record to determine potential causes for the increase in chloride load and the relative input from different sources. The chloride load at this station increased from less than 6,000 tons in water year 1982 to more than 9,000 tons in water year 2003 (fig. 17B). The average estimated chloride load from atmospheric deposition was 66 tons based on information used to create figure 3. The Quinnipiac River at Wallingford, Connecticut, had about 17 Mgal/d of wastewater discharge from three municipal wastewater-treatment facilities in 1998 (Mullaney and others, 2002); this represents about 12 percent of the average discharge for water years 1992 to 2003. In addition, the PCS database lists eight other major wastewater discharges, and four minor discharges.

The wastewater discharge is likely responsible for some of the chloride load. At a typical concentration of 91 mg/L for chloride in wastewater from sewage and animal waste (table 4), the load of chloride would be equivalent to about 2,360 tons per year, representing at least 20 percent of the load in 1998. An increase in wastewater discharge of 2.5 Mgal/d to the Quinnipiac River Basin from 1985 to 2001 was reported by Ahearn (2002). This may explain some of the increase (approximately 346 tons) in load that is apparent in figure 17B. Some of the chloride load in wastewater discharge is derived from the chloride initially in the drinking-water supply. For instance, one upstream utility in Meriden, Connecticut, reported concentrations of chloride in drinking water as large as 250 mg/L (City of Meriden, Connecticut, 2005); therefore, some chloride that initially may be derived from anthropogenic sources may be recycled.

The load of chloride from residential septic systems in the Quinnipiac River Basin (not considering systems with water softeners) was estimated by determining the number of septic systems in the basin in 1990 and then applying block-group data from the 1990 Census on the method of

Table 5. Monitoring stations where surface waters had chloride concentrations greater than 230 milligrams per liter in the glacial aquifer system, northern United States, 1991–2004.

[mg, milligram; L, liter; U, urban land use; A, agricultural land use]

Station identifier	Station name	Dominant land use	Number of chloride samples	Number of samples greater than 230 mg/L	Percent	Maximum chloride concentration (mg/L)	Months with concentration greater than 230 mg/L											
							J	F	M	A	M	J	J	A	S	O	N	D
01102500	Aberjona River at Winchester, Massachusetts	U	127	12	9.4	673	X	X	X	X								X
01356190	Lisha Kill northwest of Niskayuna, New York	U	90	7	7.8	353	X	X	X									
01391500	Saddle River at Lodi, New Jersey	U	59	1	1.7	408												X
01394500	Rahway River near Springfield, New Jersey	U	59	4	6.8	1,320	X	X		X								
03353637	Little Buck Creek near Indianapolis, Indiana	U	205	4	2.0	470	X	X	X									
04072050	Duck Creek at Seminary Road near Oneida, Wisconsin	A	93	2	2.2	425	X		X									
040869415	Lincoln Creek at 47th Street at Milwaukee, Wisconsin	U	184	42	22.8	4,330	X	X	X	X	X	X	X	X	X		X	X
04161820	Clinton River at Sterling Heights, Michigan	U	43	3	7.0	300	X										X	X
04186500	Auglaize River near Fort Jennings, Ohio	A	80	1	1.3	251										X		
05288705	Shingle Creek at Queen Avenue in Minneapolis, Minnesota	U	115	25	21.7	2,020	X	X	X								X	X
05330902	Nine Mile Creek near James Circle at Bloomington, Minnesota	U	56	7	12.5	522	X	X	X									

Table 5. Monitoring stations where surface waters had chloride concentrations greater than 230 milligrams per liter in the glacial aquifer system, northern United States, 1991–2004.—Continued

[mg, milligram; L, liter; U, urban land use; A, agricultural land use]

Station identifier	Station name	Dominant land use	Number of chloride samples	Number of samples greater than 230 mg/L	Percent	Maximum chloride concentration (mg/L)	Months with concentration greater than 230 mg/L											
							J	F	M	A	M	J	J	A	S	O	N	D
05531500	Salt Creek at Western Springs, Illinois	U	137	40	29.2	635	X	X	X	X	X							
05527800	Des Plaines River at Russell, Illinois	U	39	1	2.6	252		X										
05532000	Addison Creek at Bellwood, Illinois	U	51	16	31.4	829	X	X	X	X						X	X	X
05550500	Poplar Creek at Elgin, Illinois	U	45	12	26.7	432	X											

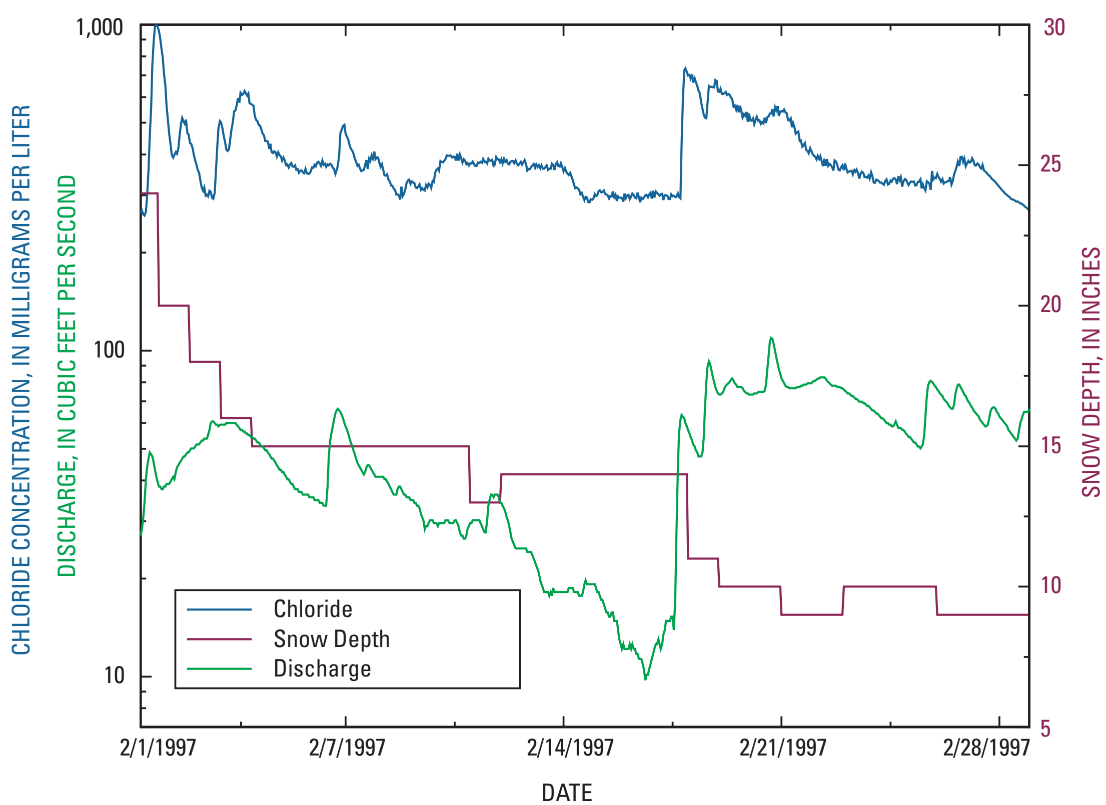


Figure 12. Chloride concentrations, snow depth, and discharge at station 05288705, Shingle Creek at Queen Avenue in Minneapolis, Minnesota, February 1997. (Data from James Fallon, U.S. Geological Survey, written commun., 2006.)

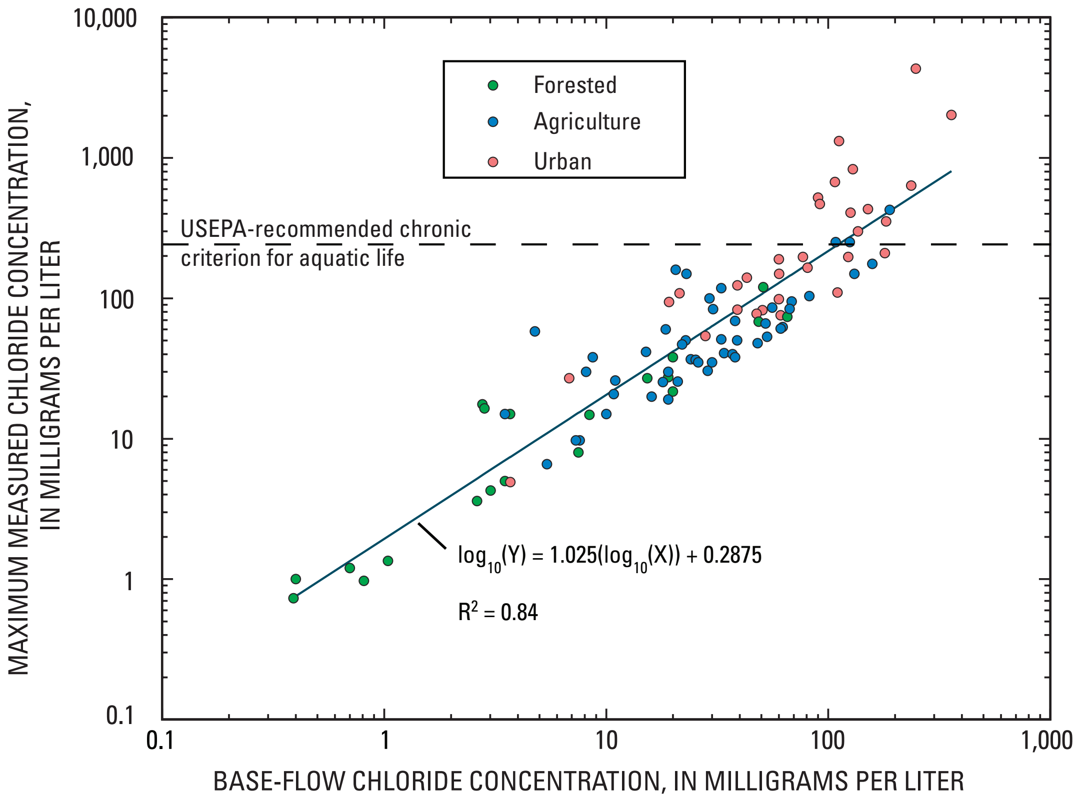


Figure 13. Chloride concentrations in base flow, plotted against maximum measured chloride concentrations at selected forested, agricultural, and urban basins in the glacial aquifer system, northern United States, 1991–2004.

Table 6. Multiple linear-regression estimates of model coefficients, standard errors, *t*-statistic and *p*-values for the dependent variable natural log of maximum measured chloride concentration in surface water for selected urban, agricultural, and forested basins, in the glacial aquifer system, northern United States, 1991–2004.

[mi, mile; mi², square mile; mm, millimeter; yr, year; <, less than; -, minus]

Variable	Units	Parameter estimate	Standard error	*t*-statistic	*p*-value
Intercept	Dimensionless	-8.3728	1.6394	-5.1071	<0.0001
Major road density	mi/mi²	1.1229	.1498	7.4941	<.0001
Potential evapotranspiration 1961–90 (effect applied only up to 625 mm/year)	mm/yr	.0212	.0028	7.4909	<.0001
Percent Dunne overland flow (saturated overland flow)	Percent of annual streamflow	-.4547	.1061	-4.2838	.0001

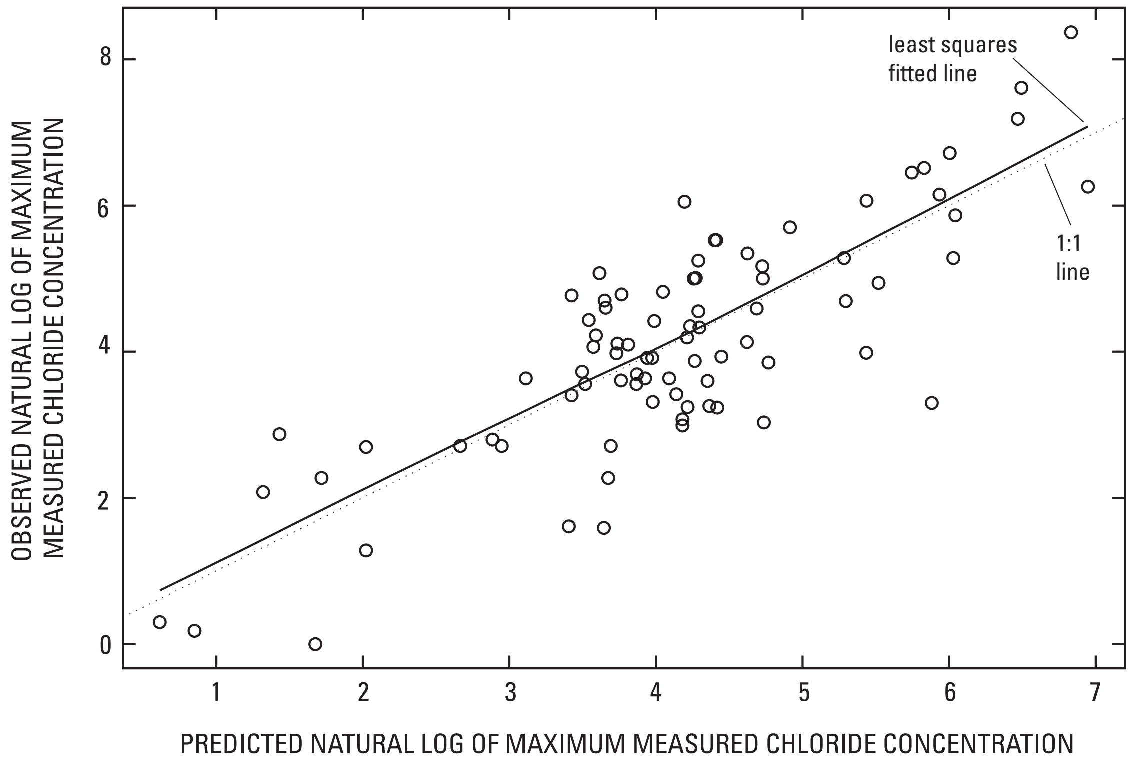


Figure 14. Predicted natural log of maximum measured chloride concentrations from multiple linear regression modeling, and observed natural log of maximum measured chloride concentrations in selected forested, agricultural, and urban basins in the glacial aquifer system, northern United States, 1991–2004.

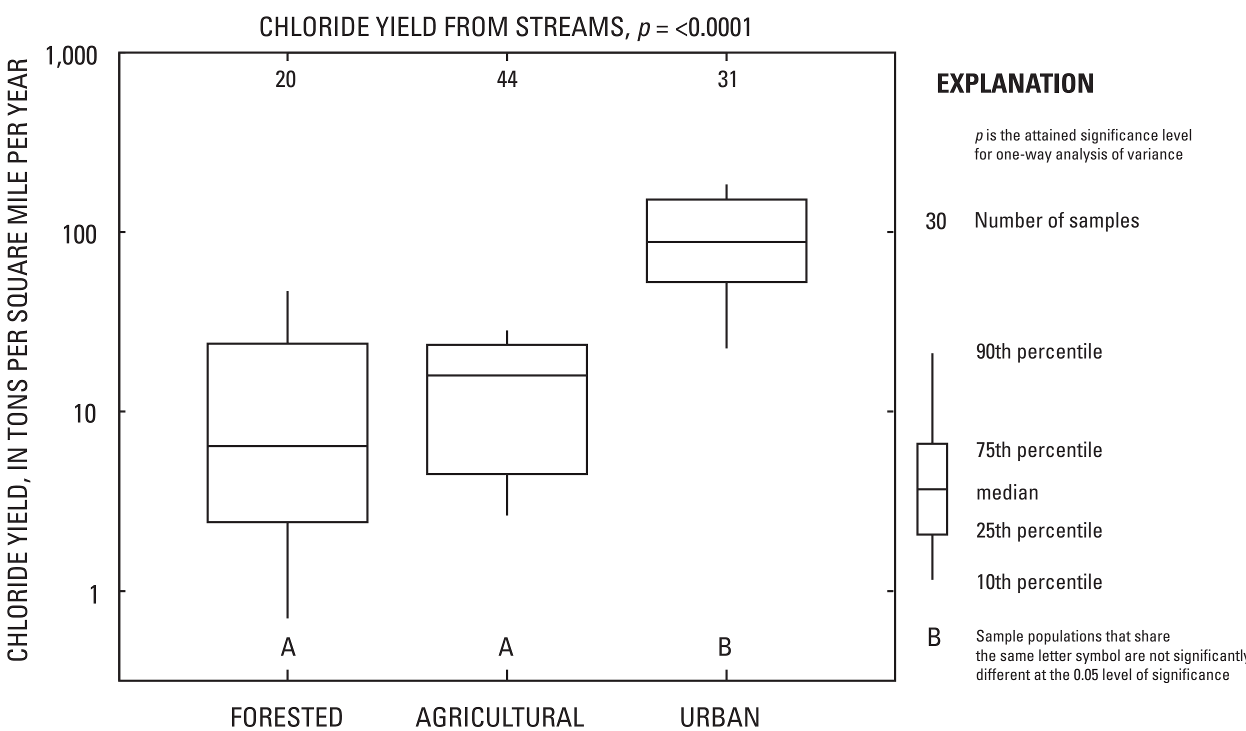


Figure 15. Distribution of annual mean chloride yields at selected surface-water-quality monitoring stations in the glacial aquifer system, northern United States, 1991–2003.

Table 7. Multiple linear regression estimates of model coefficients and standard errors, *t*-statistics, and *p*-values for the dependent variable natural log of chloride yield in selected forested, agricultural and urban basins in the glacial aquifer system, northern United States, 1991–2003.

[mi, mile; mm, millimeter; yr, year; USEPA PCS, U.S. Environmental Protection Agency Permit Compliance System; NLCDe, National Landcover Dataset (Vogelmann and others (1998), enhanced by Nakagaki and Wolock (2005)); +, plus; -, minus; <, less than]

Variable	Units	Parameter estimate	Standard error	*t*-statistic	*p*-value
Intercept	Dimensionless	-5.0085	1.0433	-4.8005	<0.0001
Highway density (U.S. numbered routes; does not include limited access highways)	mi/mi^2	2.3846	.8936	2.6687	.0093
natural log of (number of major discharges upstream of monitoring site in USEPA PCS database+1)	Dimensionless	.4149	.1809	2.2934	.0245
Potential evapotranspiration 1961–90 (effect applied only up to 700 mm/year)	mm/yr	.0123	.0017	7.2562	<.0001
Percent of urban-agricultural land (NLCDe)	Percent of basin area	.0109	.0018	5.9849	<.0001

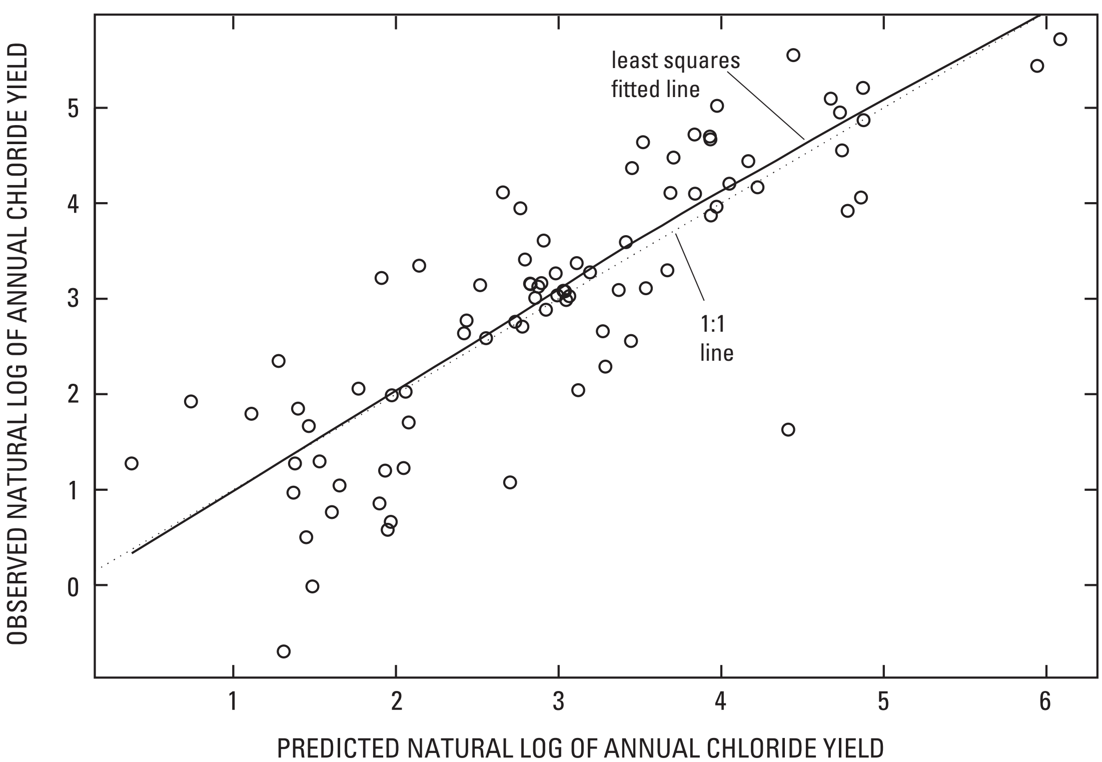


Figure 16. Predicted natural log of annual chloride yield from multiple linear regression modeling and observed natural log of chloride yield in selected forested, agricultural, and urban basins in the glacial aquifer system, northern United States, 1991–2003.

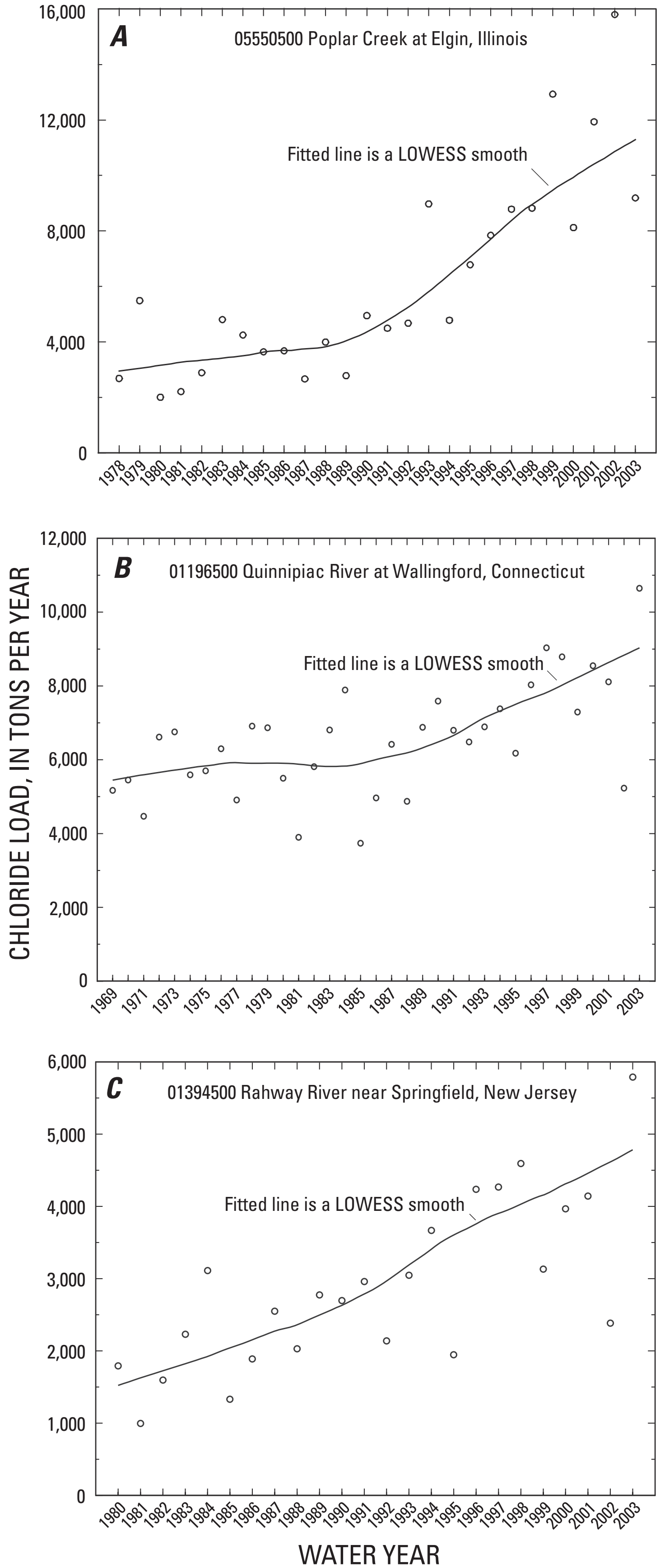


Figure 17. Chloride loads with time at selected water-quality monitoring stations in the glacial aquifer system, northern United States: *(A)* Poplar Creek at Elgin, Illinois, water years 1978–2003; *(B)* Quinnipiac River at Wallingford, Connecticut, water years 1969–2003; and *(C)* Rahway River near Springfield, New Jersey, water years 1980–2003.

sewage disposal (U.S. Department of Commerce, 1991). The estimated number of septic systems in the basin upstream from the monitoring station was 10,400. By using the estimated chloride concentration of 91 mg/L for wastewater (table 4) and assuming 300 gal/d of water use per household, the estimated load of chloride from septic systems was 430 tons in 1990. The number of septic systems likely has increased since 1990, and the presence of water softeners could increase this load substantially.

The other likely source of the increase in chloride load in this basin is the increase in the impervious area, including roads, during the period of record, and subsequent increases in deicing these areas. The impervious area of the basin above the Quinnipiac monitoring station was analyzed for 1985 and 2002 using the Impervious Surface Analysis Tool (ISAT) described by Chabaeva and others (2004). This method was used with impervious surface coefficients developed for Connecticut and population-density classes (Prisloe and others, 2003). The University of Connecticut has developed consistently interpreted satellite imagery for Connecticut land use and land cover for 1985, 1990, 1995, and 2002 (Civco and others, 1998). The ISAT method estimated that the impervious cover for the basin upstream from the monitoring station was 13.3 percent in 1985, but increased to 14.7 percent in 2002—an increase of 1.4 percent, or about 890 acres. In addition to these increases, Interstate 691 was completed in 1988, adding about 5.5 miles of four-lane road, and an estimated 85 tons of deicing salt (or about 52 tons of chloride) (Patrick Rodgers, Connecticut Department of Transportation, oral commun., 2004). Additional potential sources of chloride include 9 salt-storage areas and 20 landfills (Connecticut Department of Environmental Protection, 1995). This analysis indicates that although deicing salts may be the dominant input of chloride in many of the urban basins studied, other sources should be considered in the establishment of Total Maximum Daily Loads (TMDLs).

Summary and Conclusions

The use of salt in the United States has increased since the 1950s. Salt use has increased from 42.9 million tons in 1975 to nearly 58.5 million tons in 2005. However, the largest use of salt for many years has been the chloralkali industry. In 2005 the largest use was for deicing (39.5 percent), followed by use in the chloralkali industry (34.7 percent). The remaining uses total about 25 percent. The Northern States that compose the glacial aquifer system were the end destination for about 76 percent of the shipments of evaporated salt and rock salt.

Chloride concentrations were greater than the U.S. Environmental Protection Agency secondary maximum contaminant level (SMCL) of 250 milligrams per liter in 2.5 percent of samples from 797 shallow monitoring wells sampled (land-use wells), and in 1.7 percent of 532 drinking-water supply wells sampled from 1991 to 2003. Chloride concentrations were largest in samples from shallow monitoring wells in urban areas (median concentration of 46 milligrams per liter), followed by the concentrations in samples from agricultural areas (median, 12 milligrams per liter), and forested areas (median, 2.9 milligrams per liter).

Chloride:bromide ratios, by mass, in groundwater samples were plotted against chloride concentrations along with binary mixing curves for different sources of chloride. Samples from shallow monitoring wells in the urban areas had the largest ratios of chloride to bromide, and generally plotted on or between binary mixing curves for dilute groundwater, and (1) halite, (2) sewage and animal waste, and (3) potassium chloride fertilizer; these results indicate mixed sources of chloride. Samples that had a chloride:bromide ratio greater than 460 are dominated by anthropogenic sources of chloride. Samples that had a chloride:bromide ratio greater than 1,000 and chloride concentrations greater than 100 milligrams per liter are likely dominated by halite by mass, but may contain lesser proportions of other sources.

Samples from shallow monitoring wells in agricultural areas were not dominated by any one source of chloride, but there was a cluster of points representing samples that plotted on or near the binary mixing lines for dilute groundwater and potassium chloride or dilute groundwater and sewage and animal waste. Sources of chloride in agricultural areas include deicing salt, salt used in animal feeds, water-softening salts, salts in animal waste and fertilizer, and the use of irrigation water containing brines.

Ratios for samples from drinking-water wells indicated mixed sources of chloride, but two general patterns emerged when chloride:bromide ratios were compared by well type (private domestic drinking-water wells, and public drinking-water wells) and by region in the glacial aquifer system. The chloride:bromide ratios of the samples from the public drinking-water wells were significantly larger than for the private domestic wells, indicating anthropogenic sources of chloride. This may be a result of the integration of water of different depths and ages into public drinking-water wells due to pumping. Drinking-water-well samples from the east region of the glacial aquifer system had the highest chloride:bromide ratios followed by the central region. Samples from the west-central and west areas had low chloride:bromide ratios, indicating less human influence on water quality.

Data collected from 1991 to 2004 from 100 surface-water monitoring sites in the glacial aquifer system draining land dominated by forested land cover or urban, and agricultural land use were analyzed to compare chloride concentrations to USEPA-recommended chronic aquatic criteria, to relate maximum measured chloride concentrations to ancillary factors, and to compute annual loads so that chloride yields could be compared. Fifteen (12 urban, 3 agricultural) of the 100 sites had samples with concentrations greater than the USEPA-recommended aquatic criteria concentration for chloride of 230 milligrams per liter. Six of the 15 sites had 10 percent or more of their values greater than 230 milligrams per liter, indicating frequent occurrence of high concentrations. Concentrations of chloride greater than 230 milligrams per liter occurred most frequently during the months from November to April, indicating a likely relation with winter deicing activities.

Sites with maximum concentrations of chloride greater than 230 milligrams per liter had base-flow concentrations of chloride greater than 75 to 90 milligrams per liter. This result indicates that basins with high chloride concentrations in groundwater or wastewater discharge were more likely to exceed the recommended chronic criteria.

A multiple linear regression model was used to describe the natural log of the maximum measured chloride at 84 surface-water-monitoring stations that had ancillary data. Significant variables were the density of major roads (roads in the U.S. highway system), potential annual evapotranspiration, and the percentage of the annual streamflow derived from saturated overland flow (runoff).

The load of chloride was determined for each of 95 surface-water monitoring stations for the years with data collected from 1991 to 2003. The loads were normalized by drainage area to calculate yields of chloride. The median chloride yield was 88 tons per square mile from the basins dominated by urban land use, 15.4 tons per square mile from agricultural basins, and 6.4 tons per square mile from forested basins. The significant variables to describe chloride yields were highway density, number of major wastewater discharges, potential evapotranspiration, and percentage of urban minus agricultural land in each watershed.

Long-term historical data and the literature on this subject suggest that chloride concentrations in streams are currently increasing in urbanized and urbanizing areas, potentially exceeding recommended criteria for aquatic life now or in the coming decades. Increases in chloride load over time generally can be attributed to changes in the application of deicing salt, the expansion of road networks and impervious areas that require deicing, increases in the number of septic systems, increases in the volume of wastewater discharge, and the arrival of saline groundwater plumes from landfills and salt-storage areas over time.

Data from the Quinnipiac River in Connecticut were analyzed to explore the role of different sources of chloride in the overall chloride load. The analysis indicated that in basins with a high volume of wastewater discharge and a moderate to high density of septic systems, at least 31 percent of the chloride load can be derived from these sources. The wet deposition of chloride from precipitation represents less than one percent of the chloride load. The source of the chloride in wastewater and septic-system leachate can include chloride recycled from anthropogenic sources in the public and private water supplies; landfills and storage sites for deicing chemicals represent additional anthropogenic sources. A large part of the remaining load can be attributed to the use of deicing salt on impervious areas.

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Appendix 1

Appendix 1. Surface-water-quality monitoring stations used in analyses for chloride concentration or load estimation (or both).

[NF, North Fork; SF, South Fork; --, data not available]

USGS identifier	Station name	Hydrologic unit code	Contributing drainage area (square miles)	Dominant land use	Number of chloride samples 1992–2004	Long Term (pre-1991) data available	Maximum measured chloride concentration (milligrams per liter)	Mean chloride yield for period of selected data (tons per square mile)	Number of major permit-compliance system dischargers
01095220	Stillwater River near Sterling, MA	01070004	31.6	Forested	69		68.4	29.1	0
01101500	Ipswich River at South Middleton, MA	01090001	44.5	Urban	46		98.7	84.8	1
01102345	Saugus River at Saugus Iron Works at Saugus, MA	01090001	20.8	Urban	44		197	109.8	0
01102500	Aberjona River at Winchester, MA	01090001	24.7	Urban	127		673	182.6	1
01105000	Neponset River at Norwood, MA	01090001	34.7	Urban	32		149	88.0	0
01109000	Wading River near Norton, MA	01090004	43.3	Urban	34		77.6	64.6	2
01135300	Sleepers River (site W-5) near St. Johnsbury, VT	01080102	42.9	Agricultural	34		9.7	6.3	0
01137500	Ammonoosuc River at Bethlehem Junction, NH	01080101	87.6	Forested	34		8	5.3	0
01170100	Green River near Colrain, MA	01080203	41.4	Forested	78		15	7.3	0
01184490	Broad Brook at Broad Brook, CT	01080205	15.5	Agricultural	101		36.9	37.0	0
01184500	Scantic River at Broad Brook, CT	01080205	98.2	Agricultural	21		19	--	--
01188000	Bunnell (Burlington) Brook near Burlington, CT	01080207	4.1	Forested	52	Yes	27	25.0	0
01189000	Pequabuck River at Forestville, CT	01080207	45.8	Urban	40		76	48.1	3
01192500	Hockanum River near East Hartford, CT	01080205	73.4	Urban	129		124	60.3	2
01193500	Salmon River near East Hampton, CT	01080205	100	Forested	74	Yes	27.4	22.8	0
01196500	Quinnipiac River at Wallingford, CT	01100004	115	Urban	108	Yes	82.3	67.1	11
01199900	Tenmile River at South Dover near Wingdale, NY	01100006	192.5	Agricultural	54		35	23.6	0
01208873	Rooster River at Fairfield, CT	01100006	10.6	Urban	35		140	50.4	0
01208990	Saugatuck River near Redding, CT	01100006	21	Forested	75	Yes	38	36.2	0
01209710	Norwalk River at Winnipauk, CT	01100006	33	Urban	209	Yes	83.1	58.0	1
01349150	Canajoharie Creek near Canajoharie, NY	02020004	59.7	Agricultural	136		118	28.4	0
01356190	Lisha Kill northwest of Niskayuna, NY	02020004	15.6	Urban	90		353	103.8	0
01361200	Claverack Creek at Claverack, NY	02020006	60.6	Agricultural	24		30	--	--
01362200	Esopus Creek at Allaben, NY	02020006	63.7	Forested	55		14.8	10.5	0
01372051	Fall Kill at Poughkeepsie, NY	02020008	18.8	Urban	24		110	78.8	0
01379500	Passaic River near Chatham, NJ	02030103	100	Urban	31	Yes	210	94.7	8

Appendix 1. Surface-water-quality monitoring stations used in analyses for chloride concentration or load estimation (or both).—Continued

[NF, North Fork; SF, South Fork; --, data not available]

USGS identifier	Station name	Hydrologic unit code	Contributing drainage area (square miles)	Dominant land use	Number of chloride samples 1992–2004	Long Term (pre-1991) data available	Maximum measured chloride concentration (milligrams per liter)	Mean chloride yield for period of selected data (tons per square mile)	Number of major permit-compliance system dischargers
01380500	Rockaway River above reservoir at Boonton, NJ	02030103	116	Forested	31	Yes	120	60.7	2
01381500	Whippany River at Morristown, NJ	02030103	29.4	Urban	31	Yes	190	106.4	1
01381800	Whippany River near Pine Brook, NJ	02030103	68.5	Urban	59		165	168.6	7
01390500	Saddle River at Ridgewood, NJ	02030103	21.6	Urban	31		197	130.4	0
01391500	Saddle River at Lodi, NJ	02030103	54.6	Urban	59	Yes	408	184.1	4
01394500	Rahway River near Springfield, NJ	02030104	25.5	Urban	59	Yes	1,320	141.6	1
01435000	Neversink River near Claryville, NY	02040104	66.6	Forested	375		17.6	6.0	0
01440000	Flat Brook near Flatbrookville, NJ	02040104	64	Forested	49		21.7	20.6	0
01443500	Paulins Kill at Blairstown, NJ	02040105	126	Forested	59	Yes	73.9	61.2	2
03267900	Mad River at St. Paris Pike at Eagle City, OH	05080001	310	Agricultural	97		30.5	19.8	1
03353637	Little Buck Creek near Indianapolis, IN	05120201	17	Urban	205		470	66.9	0
03360895	Kessinger Ditch near Monroe City, IN	05120202	56.2	Agricultural	55		35	21.8	0
03366500	Muscatatuck River near Deputy, IN	05120207	293	Agricultural	28		15	9.9	0
04062085	Peshekee River near Martins Landing, MI	04030107	43.9	Forested	10		1	0.5	0
04063700	Popple River near Fence, WI	04030108	139	Forested	124		3.6	1.0	0
04071795	Pensaukee River near Krakow, WI	04030103	35.8	Agricultural	34		61	7.8	0
04072050	Duck Creek at Seminary Road near Oneida, WI	04030103	95.5	Agricultural	93		425	24.9	0
04080798	Tomorrow River near Nelsonville, WI	04030202	44	Agricultural	32		6.6	3.5	--
04085109	East River at Midway Road near de Pere, WI	04030204	47	Agricultural	26		86	--	--
040863075	North Branch Milwaukee River near Random Lake, WI	04040003	51.4	Agricultural	53		40.7	16.5	--
04086600	Milwaukee River near Cedarburg, WI	04040003	607	Agricultural	80		104	24.2	--
040869415	Lincoln Creek at 47th Street at Milwaukee, WI	04040003	9.6	Urban	184		4,330	162.9	0
04159492	Black River near Jeddo, MI	04090001	464	Agricultural	29		69.2	--	--
04161820	Clinton River at Sterling Heights, MI	04090003	309	Urban	43		300	112.1	3
04175600	River Raisin near Manchester, MI	04100002	132	Agricultural	30		25.6	17.8	0
04186500	Auglaize River near Fort Jennings, OH	04100007	332	Agricultural	80		251	26.4	3

Appendix 1. Surface-water-quality monitoring stations used in analyses for chloride concentration or load estimation (or both).—Continued

[NF, North Fork; SF, South Fork; --, data not available]

USGS identifier	Station name	Hydrologic unit code	Contributing drainage area (square miles)	Dominant land use	Number of chloride samples 1992–2004	Long Term (pre-1991) data available	Maximum measured chloride concentration (milligrams per liter)	Mean chloride yield for period of selected data (tons per square mile)	Number of major permit-compliance system dischargers
05030150	Otter Tail River near Perham, MN	09020103	335	Forested	23		5	1.8	0
05051300	Bois de Sioux River near Doran, MN	09020101	1,880	Agricultural	26		47	3.4	0
05053000	Wild Rice River near Abercrombie, ND	09020105	1,490	Agricultural	29	Yes	62.4	2.4	0
05056000	Sheyenne River near Warwick, ND	09020203	760	Agricultural	35	Yes	26	1.9	0
05058700	Sheyenne River at Lisbon, ND	09020204	2,490	Agricultural	45	Yes	48	2.8	0
05062500	Wild Rice River at Twin Valley, MN	09020108	934	Agricultural	45		15	2.2	0
05066500	Goose River at Hillsboro, ND	09020109	1,093	Agricultural	30	Yes	53.2	3.7	0
05082625	Turtle River at Turtle River State Park near Arvilla, ND	09020307	311	Agricultural	91		160	3.6	0
05085900	Snake River above Alvarado, MN	09020309	218	Agricultural	26		38	2.6	0
05112000	Roseau River below State Ditch 51 near Caribou, MN	09020314	1,420	Agricultural	29	Yes	38	1.6	0
05276005	North Fork Crow River above Paynesville, MN	07010204	232	Agricultural	35		19.9	5.5	0
05287890	Elm Creek near Champlin, MN	07010206	86	Urban	103		109	23.2	0
05288705	Shingle Creek at Queen Avenue in Minneapolis, MN	07010206	28.2	Urban	115		2,020	151.5	0
05320270	Little Cobb River near Beauford, MN	07020011	130	Agricultural	95		50.3	7.6	0
05330902	Nine Mile Creek near James Circle at Bloomington, MN	07020012	44.6	Urban	56		522	52.6	0
05331833	Namekagon River at Leonards, WI	07030002	126	Forested	32		16.4	3.3	0
05420680	Wapsipinicon River near Tripoli, IA	07080102	346	Agricultural	74		36.6	15.8	1
05451210	South Fork Iowa River northeast of New Providence, IA	07080207	224	Agricultural	100		41.6	13.3	0
05461390	Flood Creek near Powersville, IA	07080202	124	Agricultural	34		20.8	14.0	0
05464220	Wolf Creek near Dysart, IA	07080205	299	Agricultural	56		25.4	16.0	0
05520500	Kankakee River at Momence, IL	07120001	2,294	Agricultural	72	Yes	84	27.0	8
05525500	Sugar Creek at Milford, IL	07120002	446	Agricultural	75		149	15.0	0
05526000	Iroquois River near Chebanse, IL	07120002	2,091	Agricultural	33		50.3	12.9	3
05527800	Des Plaines River at Russell, IL	07120004	123	Agricultural	39		252	61.1	1
05531500	Salt Creek at Western Springs, IL	07120004	115	Urban	137		635	304.6	9
05532000	Addison Creek at Bellwood, IL	07120004	17.9	Urban	51	Yes	829	229.5	1

Appendix 1. Surface-water-quality monitoring stations used in analyses for chloride concentration or load estimation (or both).—Continued

[NF, North Fork; SF, South Fork; --, data not available]

USGS identifier	Station name	Hydrologic unit code	Contributing drainage area (square miles)	Dominant land use	Number of chloride samples 1992–2004	Long Term (pre-1991) data available	Maximum measured chloride concentration (milligrams per liter)	Mean chloride yield for period of selected data (tons per square mile)	Number of major permit-compliance system dischargers
05548105	Nippersink Creek above Wonder Lake, IL	07120006	84.5	Agricultural	38		176	51.7	1
05550500	Poplar Creek at Elgin, IL	07120006	35.2	Urban	45	Yes	432	257.1	0
05567000	Panther Creek near El Paso, IL	07130004	93.9	Agricultural	29		150	23.4	0
05568800	Indian Creek near Wyoming, IL	07130005	62.7	Agricultural	29		51	20.8	0
05572000	Sangamon River at Monticello, IL	07130006	550	Agricultural	105		95	20.3	0
05584500	La Moine River at Colmar, IL	07130010	655	Agricultural	73		60.1	14.3	1
06795500	Shell Creek near Columbus, NE	10200201	294	Agricultural	24		9.7	1.6	0
06800000	Maple Creek near Nickerson, NE	10220003	368	Agricultural	200		58.3	2.9	0
12056500	NF Skokomish River below Staircase Rapids near Hoodsport, WA	17110017	57.2	Forested	63		1.4	6.8	0
12103380	Green River above Twin Camp Creek near Lester, WA	17110013	16.5	Forested	24		1.2	3.6	0
12112600	Big Soos Creek above Hatchery near Auburn, WA	17110013	66.7	Urban	36		4.9	7.7	0
12113375	Springbrook Creek at Tukwila, WA	17110013	19	Urban	37		53.9	22.5	0
12128000	Thornton Creek near Seattle, WA	17110012	12.1	Urban	98		27	5.1	0
15241600	Ninilchik River at Ninilchik, AK	19020301	135	Forested	36		4.3	1.9	--
15266110	Kenai River below Skilak Lake Outlet near Sterling AK	19020302	1,206	Forested	36		0.1	3.0	--
15274000	SF Campbell Creek near Anchorage, AK	19020401	29.2	Forested	38		0.7	0.5	--
15275100	Chester Creek at Arctic Boulevard at Anchorage, AK	19020401	27.4	Urban	42		94.2	15.1	--
391732085414401	Clifty Creek at County Road 1150 E near Hartsville, IN	05120206	87.9	Agricultural	33		30	23.4	0
393306086585201	Big Walnut Creek at County Road 700 W at Reelsville, IN	05120203	318	Agricultural	31		40	22.0	1
394340085524601	Sugar Creek at County Road 400 S at New Palestine, IN	05120204	92.6	Agricultural	246		100	26.2	0
395355084173600	Stillwater River on Martindale Road near Union, OH	05080001	646	Agricultural	30		66.2	20.6	1
395457084095100	Great Miami River near Vandalia, OH	05080001	1,142	Agricultural	30		84.1	--	--

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