Sando and others—**Mercury and Methylmercury in Water and Bottom Sediments, Lostwood National Wildlife Refuge**—Scientific Investigations Report 2007–5219

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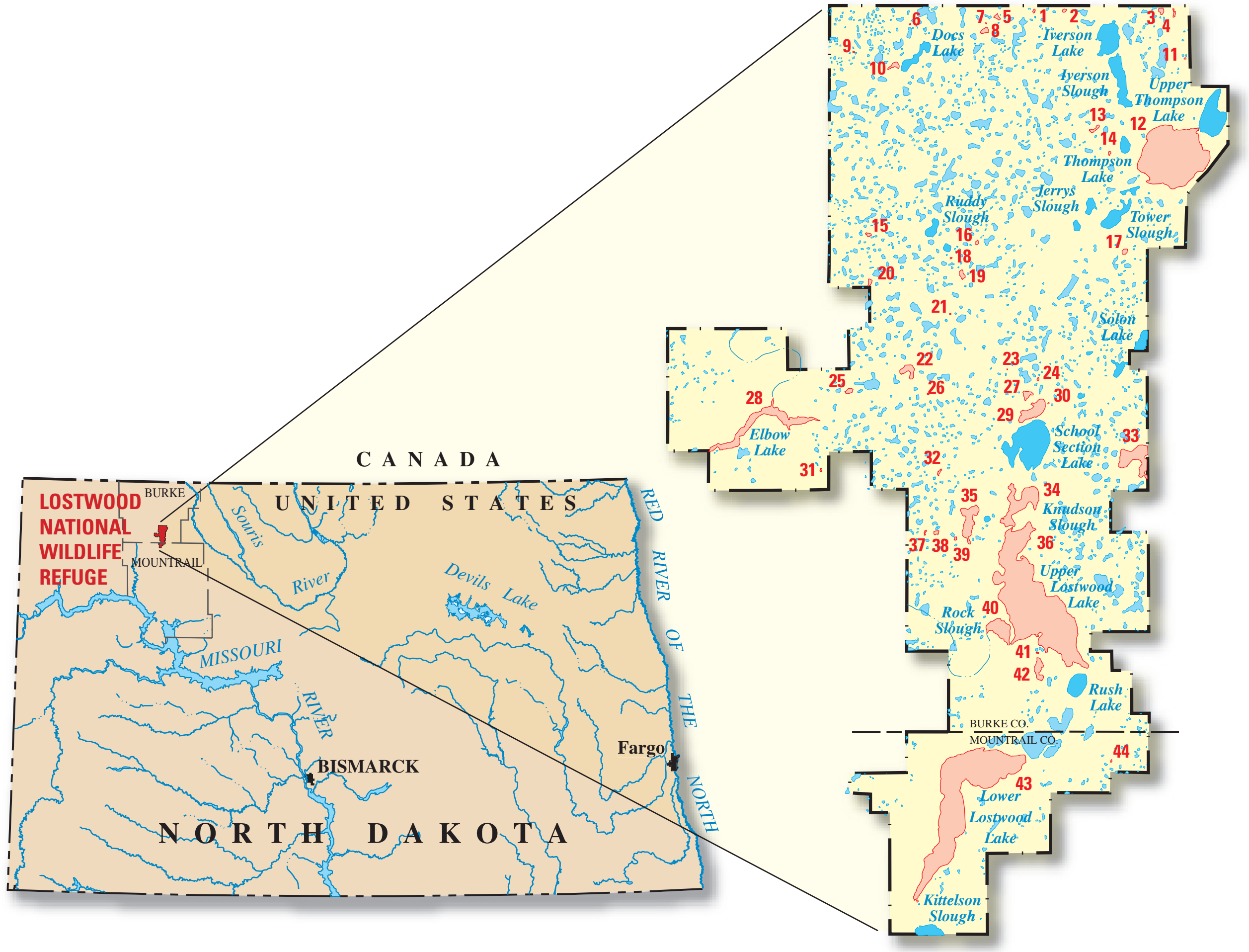
Mercury and Methylmercury in Water and Bottom Sediments of Wetlands at Lostwood National Wildlife Refuge, North Dakota, 2003–04



Scientific Investigations Report 2007–5219

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

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By Steven K. Sando, D.P. Krabbenhoft, Kevin M. Johnson, Robert F. Lundgren, and Douglas G. Emerson

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

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Conversion Factors and Datum

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	
acre	4,047	square meter (m^2)
acre	0.4047	hectare (ha)
acre	0.4047	square hectometer (hm^2)
acre	0.004047	square kilometer (km^2)
	Flow rate	
mile per hour (mi/h)	1.609	kilometer per hour (km/h)

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

°F = (1.8 x °C) + 32

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

°C = (°F-32)/1.8

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

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

Concentrations of chemical constituents in water are given in milligrams per liter (mg/L), micrograms per liter (µg/L), nanograms per liter (ng/L), or nanograms per gram (ng/g).

Mercury and Methylmercury in Water and Bottom Sediments of Wetlands at Lostwood National Wildlife Refuge, North Dakota, 2003–04

By Steven K. Sando, D.P. Krabbenhoft, Kevin M. Johnson, Robert F. Lundgren, and Douglas G. Emerson

Abstract

Certain ecosystem types, particularly wetlands, have environmental characteristics that can make them particularly sensitive to mercury inputs and that can result in large mercury concentrations in fish or other aquatic biota. To provide information needed to make effective management decisions to decrease human and wildlife exposure to methylmercury in northern prairie pothole wetlands, the U.S. Geological Survey, in cooperation with the North Dakota Department of Health, conducted a study to assess mercury and methylmercury concentrations in wetlands at the Lostwood National Wildlife Refuge (the Refuge) in northwest North Dakota. In April 2003 and 2004, water and bottom-sediment samples were collected from 44 individual wetlands that were classified as one of four wetland types. Many factors that may affect methylmercury production were considered in the study.

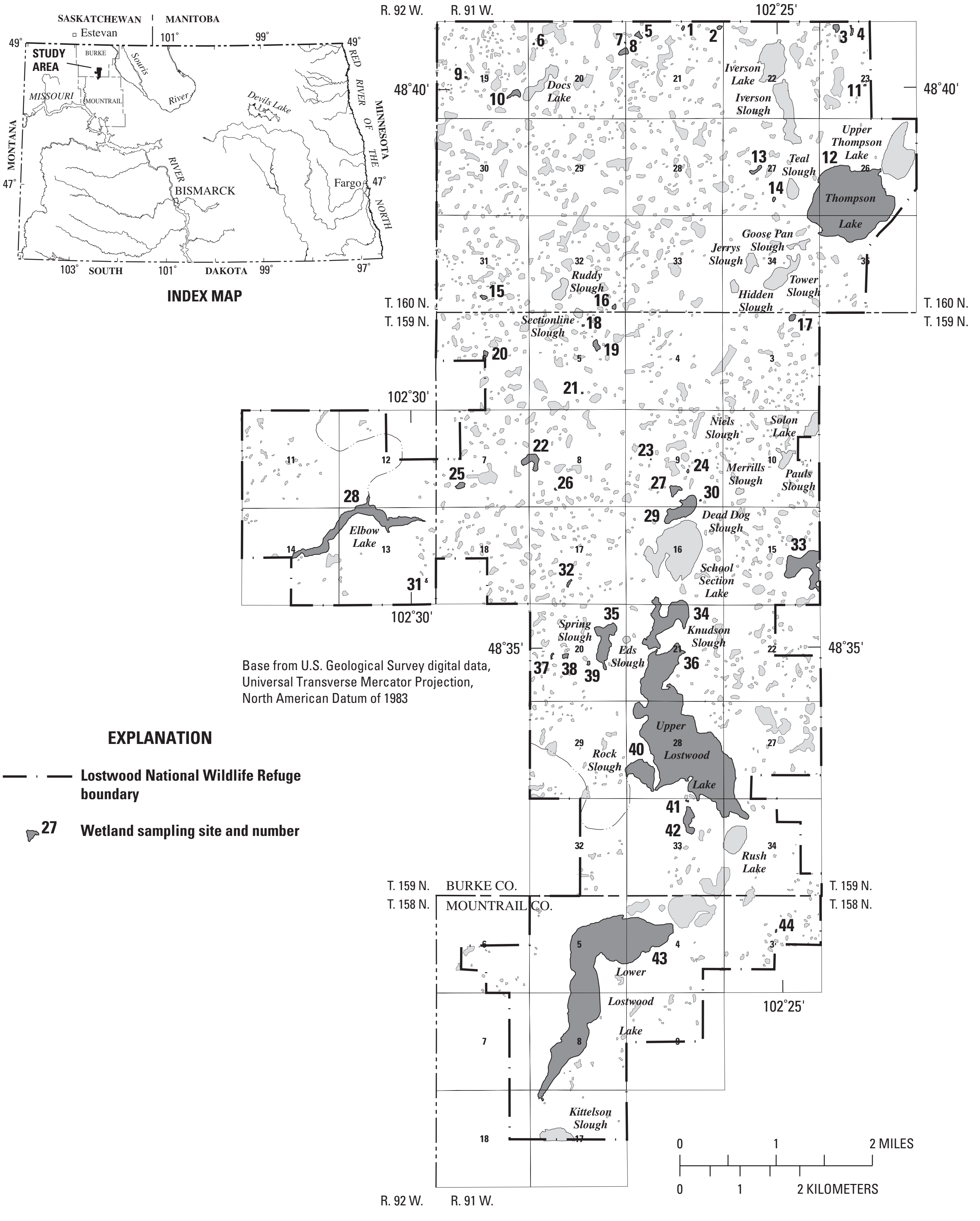
The prairie pothole wetlands at the Refuge had large ranges in major environmental characteristics. Hydrologic differences, most notably semiannual wetting and drying cycles, that are intrinsic to prairie pothole wetlands affected methylmercury concentrations. This likely resulted from the stimulation of anaerobic microbial activity following reflooding of soils, particularly soils containing substantial organic carbon. Among the four wetland types considered for this study, seasonal and semipermanent wetlands generally had the largest methylmercury concentrations. Regardless of wetland type, however, methylmercury concentrations at the Refuge are large in relation to reported concentrations for natural aquatic systems.

Introduction

In the past decade, mercury (Hg) contamination has prompted steadily increasing numbers of fish-consumption advisories. Hg-related fish-consumption advisories in 41 states now account for about 80 percent of all fish-consumption advisories in the Nation (U.S. Environmental Protection Agency, 2001). Scientific attention to the occurrence of Hg in aquatic systems has been motivated largely by concerns about effects on human health. However, aquatic food webs might be particularly vulnerable to Hg because, unlike humans, aquatic organisms cannot modify their diet in response to advisories. Widespread Hg in aquatic food webs generally is a result of anthropogenic Hg emissions (primarily from coal combustors and waste incinerators) and long-range atmospheric transport (Fitzgerald and Watras, 1989). Thus, almost all aquatic ecosystems are potentially susceptible to Hg contamination. The seemingly incongruent relation between a diffuse, low-level source of Hg (atmospheric Hg deposition) and large Hg concentrations in aquatic food webs arises from Hg methylation, which is mediated by naturally occurring sulfate-reducing bacteria. The byproduct of Hg methylation is methylmercury (MeHg), which is the most toxic and bioaccumulative form of Hg. Because MeHg readily crosses biological membranes, accumulates rapidly in exposed organisms, and can biomagnify to large concentrations in aquatic food webs, nearly all of the Hg at the top of any aquatic food web is MeHg.

Recent surveys have indicated that certain ecosystem types (for example, wetland ecosystems, newly flooded reservoirs, and seasonal impoundments) have environmental characteristics that make them particularly sensitive to Hg inputs and that can result in large Hg concentrations in fish or other aquatic biota (Spry and Wiener, 1991; Ware and others, 1991; Bodaly and others, 1997; Wiener and others, 2003). Many of these environmental characteristics are present in the prairie pothole wetlands at Lostwood National Wildlife Refuge (hereinafter referred to as the Refuge; fig. 1), and, in addition, the annual or semiannual wetting and drying cycles that are characteristic of the prairie pothole wetlands (Sloan, 1972) likely exacerbate MeHg production. Effects of wetting and drying cycles on MeHg production have been documented for the Florida Everglades (Krabbenhoft, 1996) and for Minnesota impoundments (Brigham and Krabbenhoft, 2002).

Surface-water samples collected by the U.S. Geological Survey at the Refuge in October 2002 indicate Hg methylation is pronounced. MeHg concentrations in the two water samples were 0.49 and 0.38 nanogram per liter (ng/L). These concentra-



Base from U.S. Geological Survey digital data, Universal Transverse Mercator Projection, North American Datum of 1983

Figure 1. Locations of wetland sampling sites at Lostwood National Wildlife Refuge, North Dakota.

tions are similar to those in the Florida Everglades, where large MeHg concentrations in fish have been documented (Krabbenhoft, 1996). Although concentrations in the two samples indicate prairie pothole wetlands at the Refuge are Hg sensitive and efficiently transform inorganic Hg into the much more toxic MeHg, the level of bioaccumulation that is occurring, the range in MeHg concentrations among seasons, and whether the range in physical, hydrological, and chemical characteristics of the wetlands affects MeHg production are unknown.

The many tens of thousands of prairie pothole wetlands that exist across the northern high plains serve as a critical aquatic ecosystem for native and migratory wildlife in North America (Batt and others, 1989; Swanson and Duebbert, 1989). However, little information currently (2007) is available on Hg cycling, fate, and biological exposure in the wetlands. Therefore, because environmental managers and scientists need to make effective management decisions to decrease human and wildlife exposure to MeHg, the environments that are most prone to MeHg production need to be better defined and the limiting factors for MeHg production for each ecosystem type need to be established. To provide managers and scientists with the information needed to make effective decisions, the U.S. Geological Survey, in cooperation with the North Dakota Department of Health, conducted a study to assess Hg and MeHg concentrations in prairie pothole wetlands. The purpose of this report is to describe Hg and MeHg in water and bottom sediments of wetlands at the Refuge in 2003–04. The report presents (1) analytical results of wetland water and bottom-sediment sampling; (2) investigative results of patterns in occurrence of Hg, MeHg, and other water-quality properties and constituents among wetland types and between the 2 years of sampling; and (3) investigative results of relations between Hg, MeHg, and other water-quality properties and constituents that might affect the patterns in occurrence.

Thanks are given to U.S. Fish and Wildlife Service staff at the Refuge and to Michael J. Ell and others from the North Dakota Department of Health for their cooperation and assistance during data collection. Thanks also are given to Kathy Hernandez from the U.S. Environmental Protection Agency for her support of this study. Technical reviews by Mark Brigham of the U.S. Geological Survey and James LaBaugh of the U.S. Geological Survey are greatly appreciated.

Description of Study Area

The 26,904-acre Refuge (fig. 1) was established in 1935 as a haven and breeding ground for migratory birds and other wildlife and is managed by the U.S. Fish and Wildlife Service. The Refuge is located in northwest North Dakota in Burke and Mountrail Counties, approximately 23 miles south of Canada and 70 miles east of Montana. The Refuge is the largest contiguous block of native grassland that the U.S. Fish and Wildlife Service manages in the prairie pothole region. Uplands are native mixed-grass prairie dominated by western snowberry, needlegrass, wheatgrass, grama grass, and plains muhly. Prescribed burning and grazing are the primary management tools used on the Refuge.

The Refuge contains a 5,577-acre Wilderness Area with designated Class I air quality and is listed as a Globally Important Bird Area (American Bird Conservancy, 2007). Birding is an important public use, and avian species make up the largest and most diverse faunal group on the Refuge. Shorebirds, waterfowl, raptors, and passerines use various habitats on the Refuge during migration, and the federally threatened piping plover uses shoreline habitats on the large alkali wetlands for breeding each year.

Surface runoff and subsurface seepage from rainfall and snowmelt on the Refuge results in wetlands that have a large range in size, hydrologic conditions, and water quality. Variability in climate, soils, topography, geology, and hydrology results in substantial variability in water regimes of the wetlands. The U.S. Fish and Wildlife Service (2006) used methods described by Stewart and Kantrud (1971) and Cowardin and others (1979) to classify the wetlands at the Refuge by water regime. Results of that classification were used in this study to classify each wetland sampled in the study. The wetlands were classified on the basis of water regime as one of four major types—temporary, seasonal, semipermanent, and lake. An individual wetland might actually have characteristics of more than one water regime; however, for this study, the water regime that accounted for the largest part of the total wetland area was selected to represent the wetland type. National Wetland Inventory (U.S. Fish and Wildlife Service, 2006) data indicate the Refuge contains 251 temporary wetlands that encompass 42 surface acres, 1,546 seasonal wetlands that encompass 1,211 surface acres, 152 semipermanent wetlands that encompass 700 surface acres, and 16 lake wetlands that encompass 2,040 surface acres. General qualitative characteristics of the wetland types are given in table 1.

Climate at the Refuge is semiarid. Average annual precipitation is 16.6 inches and annual temperatures typically range from -40°F to 100°F (North Dakota State Climatologist, 2004). Large seasonal and annual variability in precipitation and temperature is common; thus, annual variability in water depths and ponding periods is substantial. All wetlands, except lakes, might be completely full one year and completely dry 5 to 10 years later. Winds of 5 to 20 miles per hour are prevalent throughout most seasons, and winds of 30 to 40 miles per hour are common, particularly in spring and fall.

The primary soils on the Refuge are Zahl-Williams and Zahl-Max loams, which are characterized as thinly developed, well-drained, fine loamy soil complexes on 3- to 25-percent slopes (Soil Conservation Service, 1991). Surface runoff can be rapid on steep slopes, especially in the spring when sudden temperature increases melt snowdrifts and in the summer during intense thunderstorms.

The Refuge lies within the Missouri Coteau ecoregion (Omernik, 1987), which is a critically important rolling prairie/wetland ecosystem for resident and migratory wildlife. Armstrong (1971) provided a detailed description of the

Table 1. General qualitative characteristics of wetland types.

Wetland type	General geomorphology characteristics	General patterns in ground-water/ surface-water relations	Frequency and duration of inundation	Predominant vegetation regime
Temporary	Small in surface area and shallow	Movement of water is from wetland into local ground water throughout	Inundated for brief periods during the growing season in most years	Dense growth of grasses and sedges throughout the wetland bottom
Seasonal	Relatively small in surface area and shallow	Movement of water is from wetland into local ground water throughout	Inundated for extended periods, especially early in the growing season, in most years	Dense growth of grasses, sedges, cattails, and/or bulrushes throughout the wetland bottom
Semipermanent	Moderately large in surface area and moderately deep	Areas where movement of water is from wetland into local ground water and also areas where movement of water is from local ground water into wetland	Inundated throughout the growing season in most years and year-round in some years	Sparse growth of submergent vegetation in open-water areas; fringed with dense growth of cattails and bulrushes
Lake	Large in surface area and deep	Areas where movement of water is from wetland into local ground water but predominantly areas where movement of water is from ground water into wetland	Inundated year-round in nearly all years	Sparse to dense growth of submergent vegetation (sago pondweed and/or widgeon grass) in open-water areas; fringed with sparse growth of cattails and bulrushes

geology and topography of the Refuge area. Topographic highs consist of stagnation, dead ice moraines with 100 to 200 feet of relief. The Coteau's hummocky topography is composed of depressional (pothole) prairie wetlands that are poorly integrated by surface drainage and are principal areas of ground-water recharge. Topographic lows consist of deep stream valleys and lake depressions and are principal areas of ground-water discharge. The Refuge is overlain primarily by glacial drift that is composed mainly of till and yields little to no water. However, some drift consists of stratified glaciofluvial deposits of sand and gravel that can have varying yields. The Kenmare aquifer is composed of buried sand and gravel deposits in a valley that extends to the northeast from near Upper Lostwood Lake. Recharge to the aquifer is derived from direct precipitation, infiltration from wetlands overlying the aquifer, and inflow from adjacent undifferentiated Fort Union sediments. Discharge from ground water can occur into wetlands and small seeps at low elevations.

Large variability occurs in surface-water/ground-water interactions between individual prairie pothole wetlands (Eisenlohr and Sloan, 1968; Eisenlohr and others, 1972). For some wetlands (referred to as recharge wetlands), the movement of water primarily is from the wetlands into local ground water and water contained in the wetlands generally has had little contact with subsurface geologic materials. For other wetlands (referred to as discharge wetlands), the movement of water primarily is from ground water into the wetlands and water contained in the wetlands has had substantial contact with subsurface geologic materials. For still other wetlands (referred to as flow-through wetlands), both ground-water recharge and ground-water discharge occur and water contained in the wetlands generally has had moderate contact with subsurface geologic materials. The variability in interaction with ground water results in substantial variability in chemical characteristics of the wetlands.

Variability in climate, soils, topography, geology, and hydrology results in large differences in relative contributions of atmospheric water, surface runoff, and ground water to the wetlands. This variability also can result in substantial differences in frequency and duration of inundation among the wetlands.

Methods

Wetland Sampling Site Selection

A stratified randomization scheme was used to select wetland sampling sites for 2003. Initially, six wetlands within each of the four major wetland types were randomly selected for sampling to provide equal representation among the types and to provide a basis for statistical comparisons among the types. However, after the wetlands were sampled in April 2003, a determination was made that three of the wetlands that had been classified as semipermanent wetlands should have been classified as seasonal wetlands. Although the ability to make statistical comparisons was reduced because of the change, patterns of differences in water quality among the wetland types still were discernible. The 24 wetlands sampled in April 2003 consisted of 6 temporary wetlands, 9 seasonal wetlands, 3 semipermanent wetlands, and 6 lake wetlands.

In 2004, dry climatic conditions limited the ability to randomly select wetlands for sampling. Thus, a nonrandomized scheme was developed to select wetland sampling sites for 2004. The 25 wetlands sampled in 2004 consisted of 5 previously sampled wetlands (1 temporary wetland, 1 seasonal wetland, 1 semipermanent wetland, and 2 lake wetlands) and 20 previously unsampled wetlands (2 temporary wetlands, 9 seasonal wetlands, 7 semipermanent wetlands, and 2 lake wetlands). The five previously sampled wetlands were selected to be resampled in 2004 to provide information on interannual variability in water quality. Preliminary examination of the 2003 results indicated seasonal and semipermanent wetlands generally tended to have larger MeHg concentrations than temporary and lake wetlands (as discussed in more detail in subsequent sections of this report); therefore, seasonal and semipermanent wetlands were sampled more frequently than the other wetlands in 2004 to possibly obtain further information on Hg methylation. Also, few temporary wetlands contained adequate water for sampling in 2004.

A naming convention for individual wetland sampling sites was developed on the basis of wetland type, an arbitrary wetland identification number, the management unit containing the wetland, and the year the wetland first was sampled. Alpha codes for wetland types and management units used in the naming convention are given in table 2. For example, sampling site SS4WLD04 is seasonal wetland (SS) number 4 (4) in the Wilderness (WLD) management unit and first was sampled in 2004 (04). Water and bottom-sediment samples were collected at 44 sampling sites (table 3, fig. 1) at the Refuge in 2003 and 2004.

Table 2. Alpha codes for wetland types and management units.

Alpha code for wetland type	Alpha code for management unit
TP (Temporary)	WLD (Wilderness)
SS (Seasonal)	THL (Thompson Lake)
SM (Semipermanent)	TLS (Teal Slough)
LK (Lake)	TWS (Tower)
	ASP (Aspen)
	WDM (Windmill)
	CNC (Central Core)
	SEL (South Elbow Lake)
	GNN (Green Needle)
	FSC (Fescue)
	SSK (School Section Lake)
	KRU (Kruse)
	WWH (Western Wheat)
	WSU (West Shoreline)
	WLL (West Lower Lostwood Lake)
	ELS (South East Lower Lostwood Lake)
	ELN (North East Lower Lostwood Lake)

Table 3. Wetland sampling sites.

Sampling site number (see figure 1)	Sampling site name	U.S. Geological Survey identification number	Latitude	Longitude	Surface area (acres)	Years sampled
1	SS4WLD04	484033102261900	48°40'33"	102°26'19"	0.54	2004
2	SM4WLD04	484033102254900	48°40'33"	102°25'49"	.95	2004
3	SM1THL04	484033102241500	48°40'33"	102°24'15"	2.38	2004
4	SS1THL03	484031102240000	48°40'31"	102°24'00"	1.37	2003
5	SM3WLD04	484029102265500	48°40'29"	102°26'55"	1.79	2004
6	SS1WLD03	484025102281800	48°40'25"	102°28'18"	.17	2003
7	TP2WLD04	484024102270600	48°40'24"	102°27'06"	.09	2004
8	SM2WLD04	484020102270700	48°40'20"	102°27'07"	3.13	2004
9	TP1WLD03	484007102291300	48°40'07"	102°29'13"	.17	2003
10	SM1WLD03	483958102283400	48°39'58"	102°28'34"	3.98	2003
11	TP1THL03	484002102235000	48°40'02"	102°23'50"	.19	2003
12	LK1THL03	483859102240400	48°38'59"	102°24'04"	320.08	2003, 2004
13	SS1TLS04	483915102252200	48°39'15"	102°25'22"	2.48	2004
14	SS1TWS04	483901102250600	48°39'01"	102°25'06"	.78	2004
15	SS2WLD04	483808102290100	48°38'08"	102°29'01"	1.26	2004
16	SS3WLD04	483804102271500	48°38'04"	102°27'15"	.99	2004
17	SS1ASP04	483757102245200	48°37'57"	102°24'52"	2.28	2004
18	SS3WDM03	483754102273800	48°37'54"	102°27'38"	.25	2003
19	SM2CNC04	483743102273000	48°37'43"	102°27'30"	3.40	2004
20	SS2WDM03	483738102285800	48°37'38"	102°28'58"	1.72	2003
21	TP2CNC04	483718102274200	48°37'18"	102°27'42"	.16	2004
22	SM1WDM03	483640102282600	48°36'40"	102°28'26"	10.92	2003
23	TP1CNC03	483641102264400	48°36'41"	102°26'44"	.05	2003
24	SS3CNC04	483635102261600	48°36'35"	102°26'16"	.38	2004
25	SS4WDM03	483628102291800	48°36'28"	102°29'18"	2.80	2003
26	SS1CNC03	483626102280100	48°36'26"	102°28'01"	.24	2003
27	SM1CNC03	483626102262800	48°36'26"	102°26'28"	5.02	2003, 2004
28	LK1SEL03	483608102301100	48°36'08"	102°30'11"	77.27	2003
29	LK1CNC04	483616102261800	48°36'16"	102°26'18"	29.75	2004
30	TP1ASP03	483620102260600	48°36'20"	102°26'06"	.18	2003, 2004
31	TP1SEL03	483537102294600	48°35'37"	102°29'46"	.38	2003
32	SS1GNN04	483535102275400	48°35'35"	102°27'54"	1.11	2004
33	LK1FSC03	483541102245000	48°35'41"	102°24'50"	48.85	2003
34	LK1SSK03	483514102263300	48°35'14"	102°26'33"	68.79	2003
35	SM1KRU04	483502102272500	48°35'02"	102°27'25"	33.22	2004

Table 3. Wetland sampling sites.—Continued

Sampling site number (see figure 1)	Sampling site name	U.S. Geological Survey identification number	Latitude	Longitude	Surface area (acres)	Years sampled
36	LK1WWH03	483415102261400	48°34'15"	102°26'14"	534.75	2003
37	SS3KRU04	483457102280600	48°34'57"	102°28'06"	.60	2004
38	SS1KRU03	483456102275600	48°34'56"	102°27'56"	1.45	2003, 2004
39	SS2KRU03	483453102273500	48°34'53"	102°27'35"	.63	2003
40	LK1KRU03	483354102265300	48°33'54"	102°26'53"	34.07	2003, 2004
41	TP1WSU03	483339102261600	48°33'39"	102°26'16"	.22	2003
42	SM1WLL04	483330102261700	48°33'30"	102°26'17"	12.65	2004
43	LK1ELS04	483223102271400	48°32'23"	102°27'14"	437.52	2004
44	SS1ELN03	483229102250400	48°32'29"	102°25'04"	.29	2003

Collection, Processing, and Analysis of Water and Bottom-Sediment Samples

For each year of data collection, wetlands were sampled about 2 weeks after icemelt and snowmelt when wetland water levels would be expected to be near annual maxima and water temperatures would have increased enough to increase biological metabolic rates. Wetlands were sampled during April 21–24, 2003, and during April 26–28, 2004.

For a given wetland, water samples were collected from the open-water part of the wetland at a single location beyond the fringe of emergent vegetation. Because the Refuge wetlands generally are shallow (typically less than 5 feet deep) and tend to be well mixed by wind-generated turbulence, a single sampling location for each wetland was assumed to be representative of the entire wetland. Collection of water samples at a single location also reduced the potential for contamination of water samples analyzed for Hg and MeHg. Because Hg and MeHg both occur in extremely small concentrations in natural waters, samples can be affected by contamination artifacts from inadequately cleaned sample containers, unintentional direct contact by sampling crews, and the environment (for example, airborne particles). For this study, two-person, ultra-clean sampling procedures (Olson and DeWild, 1999) were used to collect all water and bottom-sediment samples.

Samples were analyzed for the properties and constituents given in table 4. Because of the ultra-clean sampling procedures needed for the collection of Hg and MeHg water samples, these samples were collected first and then non-Hg water samples and bottom-sediment samples were collected. Field measurements were made after all samples were collected. Water samples were collected using the hand grab method from about mid-depth in the water column. Sample bottles were rinsed three times with wetland water before final filling. Field crews wore clean TYVEK suits, shoulder-length polyethylene gloves, and wrist-length latex gloves. Fluorocarbon polymer sample bottles used for collection of Hg water samples were contained in double polyethylene bags. Any materials and containers that contacted the water samples were rigorously cleaned using procedures described by Olson and DeWild (1999), DeWild and others (2002), and the U.S. Environmental Protection Agency (2002). Sample-collection procedures and the types of bottles used for collection of major-ion, suspended-solids, non-Hg trace-element, and organic-carbon water samples followed standard protocols (Wilde and others, 1998; North Dakota Department of Health, 2003a and 2003b).

Bottom-sediment samples were collected from depositional zones where the surficial sediments consisted primarily of fine-grained material. Samples were collected with plastic scoops or polycarbonate cylinders that were rigorously cleaned using procedures described by Olson and DeWild (1999), DeWild and others (2002), and the U.S. Environmental Protection Agency (2002). Sampling equipment was rinsed three times with wetland water before sample collection. For each wetland, subsamples of the top 1 to 2 inches of sediment were collected from 5 to 10 locations within a radius of about 30 feet from where the water sample was collected. The subsamples were composited into double polyethylene bags and homogenized, which resulted in a sample of about 1 to 2 liters of sediment material for each wetland. A single sediment sample then was taken from the composited/homogenized material, placed

Table 4. Properties and constituents for which water samples and bottom-sediment samples were analyzed.

Property or constituent	Study reporting level and units
Field measurements	
Depth to bottom from water surface at sampling site location	0.1 foot[1]
Barometric pressure	1 millimeter mercury[1]
Specific conductance	5 microsiemens per centimeter[1]
pH	0.1 standard unit[1]
Air temperature	0.1 degree Celsius[1]
Water temperature	0.1 degree Celsius[1]
Turbidity	1 formazin nephelometric unit[1]
Dissolved oxygen	0.1 milligram per liter[1]
Laboratory analyses **Major ions, suspended solids, and selected trace elements** **(North Dakota Department of Health Laboratory, Bismarck, North Dakota)**	
Hardness, total	1 milligram per liter as calcium carbonate[2]
Calcium, dissolved	2 milligrams per liter[3]
Magnesium, dissolved	1 milligram per liter[3]
Sodium, dissolved	3 milligrams per liter[3]
Percent sodium, in equivalents of major cations	1 percent[2]
Sodium adsorption ratio	0.1[2]
Potassium, dissolved	1 milligram per liter[3]
Alkalinity, whole-water	1 milligram per liter as calcium carbonate[3]
Bicarbonate, whole-water	1 milligram per liter[3]
Carbonate, whole-water	1 milligram per liter[3]
Hydroxide, whole-water	1 milligram per liter[3]
Sulfate, dissolved	0.3 milligram per liter[3]
Chloride, dissolved	0.3 milligram per liter[3]
Solids, dissolved, sum of constituents	1 milligram per liter[2]
Solids, suspended	5 milligrams per liter[3]
Aluminum, dissolved	50 micrograms per liter[3]
Antimony, dissolved	1 microgram per liter[3]
Arsenic, dissolved	1 microgram per liter[3]
Barium, dissolved	1 microgram per liter[3]
Beryllium, dissolved	1 microgram per liter[3]
Boron, dissolved	50 micrograms per liter[3]
Cadmium, dissolved	1 microgram per liter[3]
Chromium, dissolved	1 microgram per liter[3]
Copper, dissolved	1 microgram per liter[3]
Iron, dissolved	10 micrograms per liter[3]
Lead, dissolved	1 microgram per liter[3]
Manganese, dissolved	10 micrograms per liter[3]
Nickel, dissolved	1 microgram per liter[3]
Selenium, dissolved	1 microgram per liter[3]
Silver, dissolved	1 microgram per liter[3]
Thallium, dissolved	1 microgram per liter[3]
Zinc, dissolved	1 microgram per liter[3]

Table 4. Properties and constituents for which water samples and bottom-sediment samples were analyzed.—Continued

Property or constituent	Study reporting level and units
Mercury and organic carbon (U.S.Geological Survey Wisconsin Water Science Center Mercury Laboratory, Madison, Wisconsin)	
Methylmercury, water, dissolved	0.04 nanogram per liter[4]
Methylmercury, water, suspended	0.03 nanogram per liter[5]
Methylmercury, water, whole-water	0.04 nanogram per liter[2]
Total mercury, water, dissolved	0.1 nanogram per liter[6]
Total mercury, water, suspended	0.1 nanogram per liter[7]
Total mercury, water, whole-water	0.1 nanogram per liter[2]
Organic carbon, water, dissolved	0.01 milligram per liter[8]
Organic carbon, water, whole-water	0.1 milligram per liter[8]
Methylmercury, bottom sediment (methylmercury, solid, total, dry weight)	0.4 nanogram per gram[5]
Total mercury, bottom sediment (mercury, solid, total, dry weight)	0.01 nanogram per gram[7]
Loss on ignition, bottom sediment (loss on ignition, solids)	1 percent[9]

[1] Wilde and others, 1998.

[2] Calculated.

[3] North Dakota Department of Health, 2003a, 2003b.

[4] DeWild and others, 2002.

[5] DeWild and others, 2004.

[6] Olson and DeWild, 1997a.

[7] Olund and others, 2004.

[8] Olson and DeWild, 1997b.

[9] American Public Health Association, American Water Works Association, and Water Pollution Control Federation, 1975.

in a fluorocarbon polymer vial, and double bagged in zip seal polyethylene bags.

Field measurements were determined for each wetland by wading into the open-water part of the wetland beyond the fringe of emergent vegetation and making in-situ measurements. Field measurements were made at about mid-depth in the water column.

Immediately after collection, Hg water samples were placed on ice and transported to a clean processing area at the Refuge headquarters. Hg and MeHg water samples were filtered (if needed) and acidified using procedures described by Wilde and others (1998) and DeWild and others (2002). Major-ion, suspended-solids, trace-element, and organic-carbon water samples were processed using standard protocols for those constituents (Wilde and others, 1998; North Dakota Department of Health, 2003b) and analyzed by the North Dakota Department of Health Laboratory (North Dakota Department of Health (2003b). Hg, MeHg, and organic-carbon water and bottom-sediment samples were analyzed by the U.S. Geological Survey Wisconsin Water Science Center Mercury Laboratory, a class-100 clean-room facility.

In this report, the term total with respect to Hg refers to the analytical results for a sample processed using U.S. Environmental Protection Agency Method 1631 (Olson and DeWild, 1999). Method 1631 entails rigorous digestion and oxidation steps that render all Hg species in a sample (inorganic and organic forms) to the mercuric ion state (Hg^{+2}). Following digestion and oxidation, the sample is treated with stannous chloride ($SnCl_2$), which serves to chemically convert all Hg to gaseous Hg^0, which is sparged from the sample and detected by Atomic Fluorescence Spectroscopy. MeHg samples were analyzed using procedures described by DeWild and others (2002). These procedures involve a two-part process of distillation and ethylation followed by chromatographic separation of the organic Hg forms (both mono- and dimethylmercury) and detection by Atomic Fluorescence Spectroscopy. For this study, all organic Hg present in the water samples was assumed to be monomethylmercury because dimethylmercury is exceedingly rare and has been detected only in marine settings. Inorganic Hg concentrations in water samples were calculated by subtracting MeHg concentrations from total Hg (THg) concentrations. In this calculation, if either the MeHg or THg concentration for a given sample was less than the study reporting level (table 4), the concentration for the less-than fraction was assumed to be zero. To determine dissolved concentrations of Hg and organic-carbon constituents, water samples were passed through ultra-clean, 0.7-micrometer-nominal-pore size, quartz-fiber filters in the processing area at the Refuge headquarters. Thus, in this report, the term dissolved with respect to Hg and organic-carbon constituents is operationally defined as that part of a water sample that passes through a 0.7-micrometer filter, noting that some fraction of the dissolved part is composed of filter-passing colloids. To determine suspended concentrations of Hg constituents, known volumes of sample water were passed through ultra-clean, 0.7-micrometer-nominal-pore-size, quartz-fiber filters. The filter and retained suspended material were digested and the resulting solution was analyzed using methods described by Olund and others (2004) for Hg and by DeWild and others (2004) for MeHg. Mass/volume suspended concentrations of Hg constituents then were determined on the basis of the original known volume of sample water that was filtered. To determine whole-water concentrations of Hg constituents, dissolved and suspended concentrations were summed. If either the dissolved or suspended concentration for a given sample was less than the study reporting level, the concentration for the less-than fraction was assumed to be zero. To determine dissolved concentrations of major-ion and non-Hg trace-element constituents, water samples were passed through clean 0.45-micrometer-nominal-pore-size, membrane filters at the North Dakota Department of Health Laboratory. Thus, in this report, for major-ion and non-Hg trace-element constituents, the term dissolved is operationally defined as that part of a water sample that passes through a 0.45-micrometer filter.

Collection, Processing, and Analysis of Quality-Assurance/Quality-Control Samples

Quality-assurance/quality-control (QA/QC) samples were collected to assess precision and accuracy of collected data. Two field-equipment blank samples (water samples only) and two replicate samples (water and bottom-sediment samples) were collected in both 2003 and 2004 and analyzed for laboratory analytical constituents (table 4). Thus, a total of four field-equipment blank samples and four replicate samples were collected. This number of QA/QC samples represents about 16.3 percent of the number of environmental samples collected. Detailed information on methods used to collect, process, and analyze the blank and replicate samples is given in supplement 1 along with results of the statistical analyses.

Climatic Conditions Prior to Sample Collection

Climatic conditions prior to sample collection in 2003 and 2004 (table 5, fig. 2) differed substantially and likely affected the results obtained during this study. Effects of the differences on wetland inundation in the northern part of the Refuge in the springs of 2003 and 2004 are shown in figure 3. For the area of the Refuge (fig. 3), the number of inundated wetlands in 2004 was about 30 percent smaller than the number in 2003 and the total wetted surface area in 2004 was about 35 percent smaller than the total in 2003. These data indicate substantially less runoff of meltwater to the wetlands in 2004 than in 2003. The largest decreases between 2003 and 2004 in both the number of inundated wetlands and the total wetted surface area occurred for temporary, seasonal, and semipermanent wetlands. In the prairie pothole region, in years with little winter snowpack and dry antecedent moisture conditions, contribution of water to wetlands generally is from melting snow and ice on the wetland water surface or within the localized wetland depression, and the amount of runoff from surrounding terrestrial areas with associated flushing of soils is small (Waiser and Robarts, 2004).

The climatic data collected at a National Weather Service station located at Kenmare, North Dakota (about 15 miles northeast of the Refuge; fig. 3), are presented to provide additional information on climatic conditions at the Refuge during the study period. References to average conditions refer to conditions during 1971–2000. During the year prior to sample collection in late April 2003, total precipitation in the Refuge area was above average and mean air temperature was near average. The summer of 2002 was relatively cool and wet and soil moisture probably was near or above average prior to frozen soil conditions in winter. During the winter of 2002–03, mean air temperature was near average and snowfall was much above average; more than half of the winter snowfall occurred after February 1. The first week of April 2003 was cold and the area received about 2 inches of precipitation as snow. Air temperatures rose abruptly in the second week of April and were consistently above freezing until the time of sample collection (April 21–24). These climatic conditions resulted in rapid melting of the snowpack and substantial runoff to the Refuge wetlands.

During the year prior to sample collection in late April 2004, total precipitation in the Refuge area was near average and mean air temperature was near average. However, the summer of 2003 was extremely hot and dry. The Refuge wetlands were either dry or contained below-average water volumes, and soil moisture probably was substantially depleted prior to frozen soil conditions in winter. During the winter of 2003–04, mean air temperature was below average and snowfall was much above average. However, most of the winter snowfall occurred during November–January. February–April precipitation was much below average. These climatic conditions probably resulted in degradation of the snowpack by sublimation and evaporation in late winter. In late March, air temperatures

Table 5. Climatic conditions at Kenmare, North Dakota (located about 15 miles northeast of Lostwood National Wildlife Refuge), prior to sample collection in 2003 and 2004.

Time period		Mean daily air temperature (degrees Fahrenheit)	Exceedance percentile[1] of mean daily air temperature for specified time period (based on comparison with 1971–2000 data)	Total precipitation (inches)	Exceedance percentile[1] of total precipitation for specified time period (based on comparison with 1971–2000 data)
Climatic conditions prior to sample collection in 2003	May 1, 2002, to April 30, 2003	38.9	60	18.8	32
	June 1, 2002, to October 31, 2002	58.3	60	11.2	37
	November 1, 2002, to April 30, 2003	21.1	54	6.2	22
	February 1, 2003, to April 30, 2003	23.0	80	3.5	23
Climatic conditions prior to sample collection in 2004	May 1, 2003, to April 30, 2004	40.0	46	16.8	54
	June 1, 2003, to October 31, 2003	61.1	6	8.0	77
	November 1, 2003, to April 30, 2004	20.0	64	6.1	22
	February 1, 2004, to April 30, 2004	28.0	46	1.4	83

[1] Exceedance percentile indicates the percentage of time the specified value is equaled or exceeded on average.

were consistently above freezing; however, they did not reach the same levels that occurred just prior to sample collection in April 2003. The mean daily air temperature during March 15–April 15, 2004, was 34.7°F. The relatively small snowpack that remained in April 2004 probably melted relatively slowly, allowing substantial infiltration to soils that had been substantially depleted of moisture during the hot, dry summer of 2003. Thus, climatic conditions prior to sample collection in 2004 probably resulted in little runoff to the Refuge wetlands.

Mercury and Methylmercury in Water and Bottom Sediments

Analytical results of wetland water and bottom-sediment sampling; patterns in the occurrence of Hg, MeHg, and other water-quality properties and constituents among wetland types and between the 2 years of sampling; and relations between Hg, MeHg, and other water-quality properties and constituents that might affect the patterns in occurrence are discussed in this section. Data are presented to document patterns in field data and major-ion, non-Hg trace-element, and organic-carbon constituents. The distinct patterns in the occurrence of non-Hg constituents in the Refuge wetlands are associated with distinct patterns in the occurrence of Hg and MeHg. The relations between some of the non-Hg constituents are discussed with a focus on how they might affect the occurrence of Hg and MeHg in the Refuge wetlands.

Wetland Water and Bottom-Sediment Sampling

Field measurements were made during this study and primary environmental samples were collected to provide information on wetland water and bottom-sediment quality. QA/QC samples were collected to document that the data were of sufficient quality to satisfy study objectives. Analytical results for QA/QC samples are given in supplements 1 through 6. Analytical results for primary environmental samples are given in supplement 2 (water-quality property and constituent values), supplement 3 (major-ion, suspended-solids, and non-Hg trace-element data for water samples), and supplement 4 (Hg and organic-carbon data for water and bottom-sediment samples).

In general, results for field-equipment blank samples indicate the overall process of sample collection, processing, and analysis did not introduce substantial contamination and did not substantially bias study results. Further, results for replicate samples generally indicate the overall process of sample collection, processing, and analysis provided accurate precision and did not substantially bias study results.

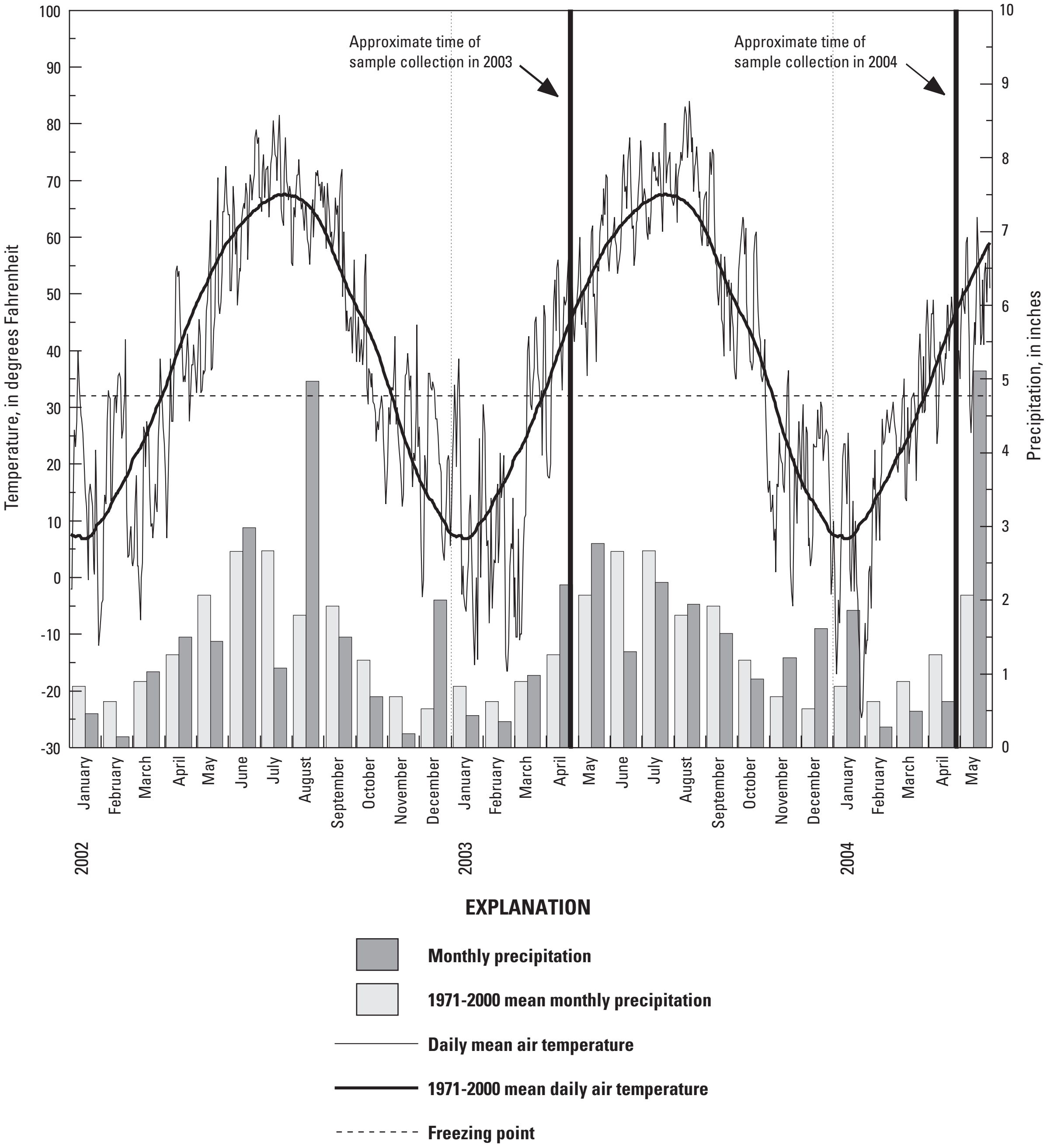
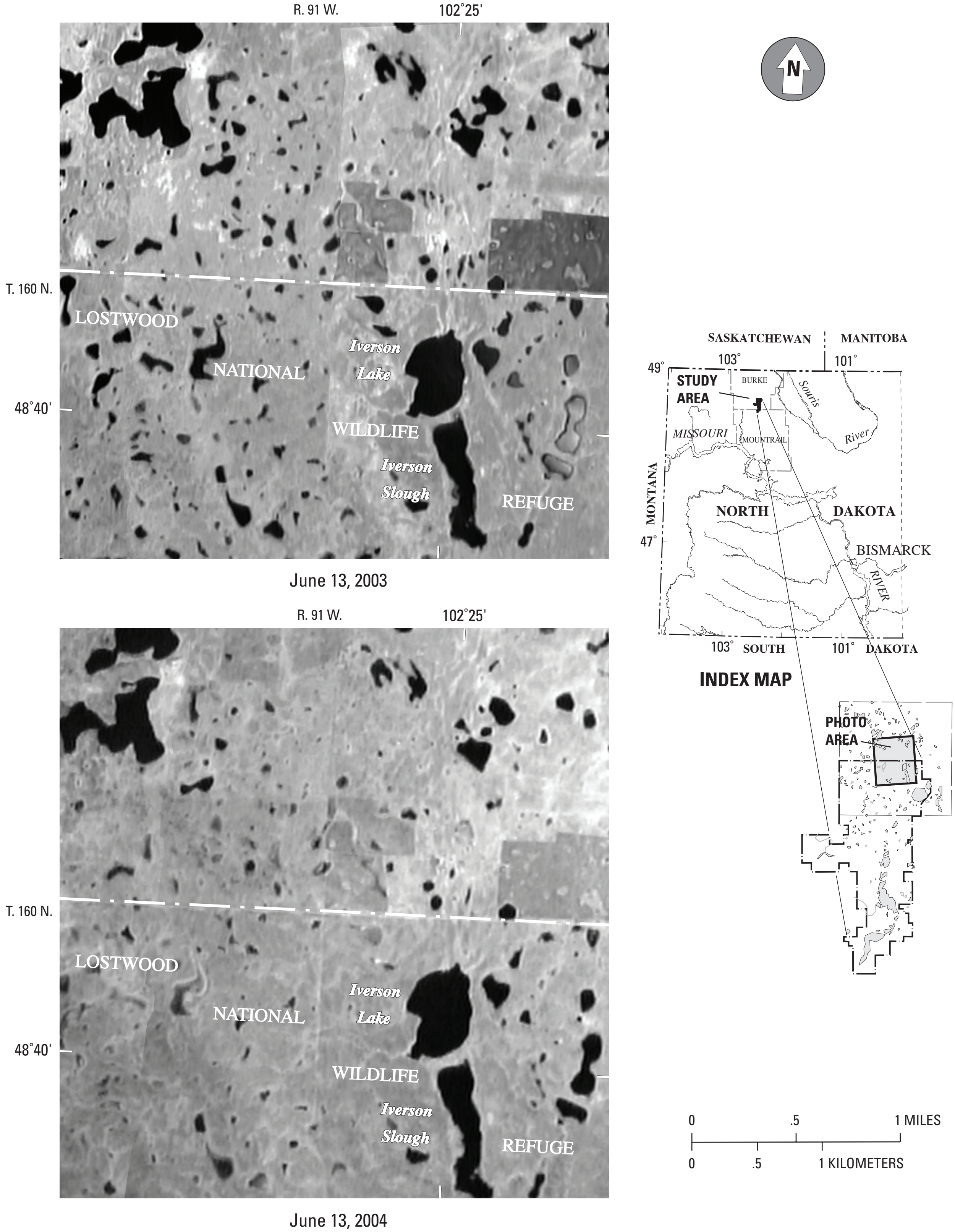


Figure 2. Climatic data collected at Kenmare, North Dakota (located about 15 miles northeast of Lostwood National Wildlife Refuge), for January 2002 through May 2004 (High Plains Regional Climate Center, 2007).



June 13, 2003

June 13, 2004

Figure 3. Differences in wetland inundation between 2003 and 2004 for the northern part of Lostwood National Wildlife Refuge, North Dakota.

Patterns in Occurrence of Mercury, Methylmercury, and Other Water-Quality Properties and Constituents

Mercury Constituents in Water and Bottom-Sediment Samples and Properties of Bottom-Sediment Samples

Most MeHg, inorganic mercury, and THg in the Refuge wetlands was dissolved (supplement 4). Three-fourths of the water samples collected had dissolved fractions of both MeHg and THg that accounted for about 70 percent or more of the whole-water concentrations of those constituents. Hg in the dissolved state commonly is predominant in surface waters that contain appreciable amounts of dissolved organic carbon (DOC), which is the dominant complexing agent for Hg and MeHg in most natural waters (Krabbenhoft and others, 1999; Wiener and others, 2003). Because MeHg, inorganic mercury, and THg were predominantly dissolved, dissolved and whole-water concentrations of those constituents showed similar patterns among wetland types and between years (supplement 4). Thus, although analytical results are presented for dissolved, suspended, and whole-water concentrations of MeHg, inorganic mercury, and THg, discussion summarizing differences among wetland types and between years generally is limited to whole-water concentrations that are predominantly in the dissolved state. Statistical summaries of concentrations of selected Hg constituents in water and bottom-sediment samples and percent loss on ignition for bottom-sediment samples are shown in figures 4 and 5.

Whole-water MeHg concentrations ranged from less than 0.04 ng/L for site 43 (LK1ELS04) for 2004 to 9.56 ng/L for site 38 (SS1KRU03) for 2003 (supplement 4). Most whole-water MeHg concentrations (the middle 80 percent) were between 0.11 and 1.62 ng/L (fig. 4). Generally, MeHg concentrations in the Refuge wetlands were large in relation to reported concentrations for natural aquatic systems (Bloom, 1989; Krabbenhoft and others, 1999; Ullrich and others, 2001; Wiener and others, 2003). Bloom (1989) reported that MeHg concentrations in natural surface waters typically range from 0.02 to 0.1 ng/L. For 2003, the median whole-water MeHg concentration for lake wetlands (0.17 ng/L) was less than that for temporary wetlands (0.53 ng/L) which was less than that for seasonal wetlands (1.09 ng/L) which was less than that for semipermanent wetlands (1.24 ng/L) (fig. 4). For 2004, the median whole-water MeHg concentration for lake wetlands (0.16 ng/L) was less than that for temporary and semipermanent wetlands (0.28 ng/L) which was less than that for seasonal wetlands (0.32 ng/L) (fig. 4). Within types, median whole-water MeHg concentrations were larger for 2003 than for 2004 for temporary, seasonal, and semipermanent wetlands and were about equal for 2003 and 2004 for lake wetlands. For wetlands sampled in both 2003 and 2004, whole-water MeHg concentrations were larger for 2003 than for 2004 except for site 40 (LK1KRU03) where the concentrations were about equal for 2003 and 2004.

MeHg concentrations in the Refuge wetlands in general, and in the seasonal and semipermanent wetlands in particular, were substantially larger in 2003 than in 2004. Hg can be extensively processed in natural aquatic systems, and all data needed to assess in detail various factors that might have affected differences in MeHg concentrations between years were not collected during this study. However, differences in MeHg concentrations in the wetlands between 2003 and 2004 might be attributable to one or more of several factors, including (1) substantial differences in contributions of inorganic Hg and organic carbon (the processing of which is critical to Hg methylation) to the wetlands between years; (2) substantial differences in water chemistry of the wetlands between years that affected the processing of Hg and, thus, the fate and concentrations of different forms of Hg in the wetlands; and (3) differences in both contributions of inorganic Hg/organic carbon and water chemistry between years that affected the production of MeHg in the wetlands between years.

Whole-water inorganic Hg concentrations ranged from 0.61 ng/L for site 3 (SM1THL04) for 2004 to 11.7 ng/L for site 42 (SM1WLL04) for 2004 (supplement 4). Most whole-water inorganic Hg concentrations (the middle 80 percent) were between 1.35 and 7.79 ng/L (fig. 4). Generally, inorganic Hg concentrations in the Refuge wetlands were within ranges reported for natural aquatic systems (Krabbenhoft and others, 1999; Wiener and others, 2003). For 2003, the median whole-water inorganic Hg concentration for lake wetlands (1.41 ng/L) was less than that for temporary wetlands (3.82 ng/L) which was less than that for seasonal wetlands (5.47 ng/L) which was less than that for semipermanent wetlands (5.50 ng/L) (fig. 4). For 2004, the median whole-water inorganic Hg concentration for lake wetlands (2.10 ng/L) was less than that for semipermanent wetlands (3.20 ng/L) which was less than that for seasonal wetlands (3.84 ng/L) which was less than that for temporary wetlands (6.20 ng/L) (fig. 4). Within types, median whole-water inorganic Hg concentrations were smaller for 2003 than for 2004 for temporary and lake wetlands and were larger for 2003 than for 2004 for seasonal and semipermanent wetlands. For wetlands sampled in both 2003 and 2004, whole-water inorganic Hg concentrations were larger for 2003 than for 2004 for site 38 (SS1KRU03) and site 27 (SM1CNC03), were about equal for 2003 and 2004 for site 30 (TP1ASP03) and site 40 (LK1KRU03), and were smaller for 2003 than for 2004 for site 12 (LK1THL03).

Although variability in inorganic Hg concentrations occurred between years, the differences in concentrations were relatively small compared to differences in MeHg concentrations (fig. 4). This pattern might suggest that, although inorganic Hg concentrations generally were similar between years, the inorganic Hg was not as bioavailable in 2004 as in 2003 and/or other conditions for methylation of inorganic Hg were more suitable in 2003 than in 2004. Although the inorganic Hg concentrations might have been generally similar between years, the contributions of inorganic Hg to the wetlands may not have been similar between years. The total volume of water contained in the Refuge wetlands probably was substantially smaller in 2004 than in 2003. Thus, the total mass of inorganic Hg in the Refuge wetlands was substantially smaller in 2004

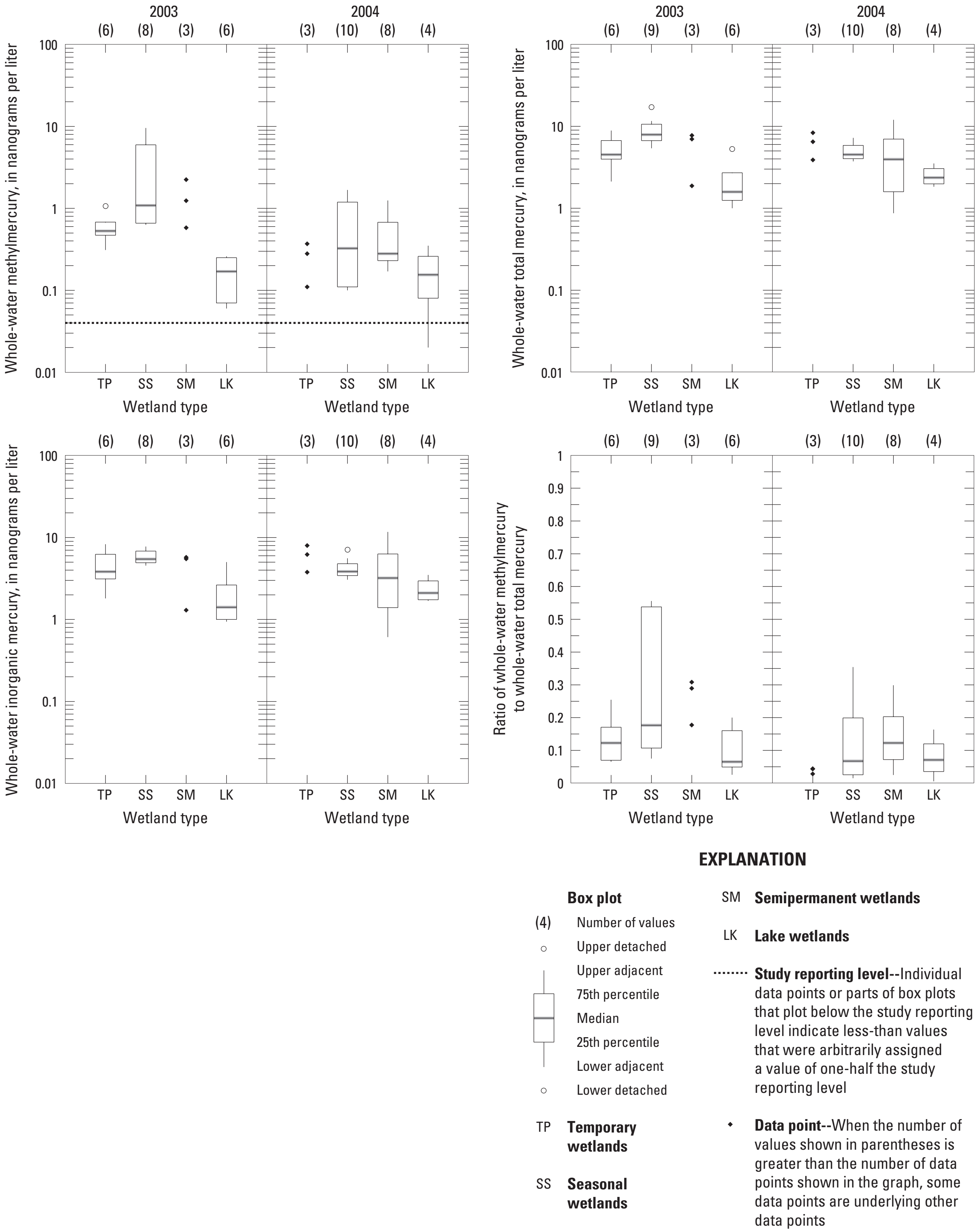


Figure 4. Concentrations of selected mercury constituents in water samples.

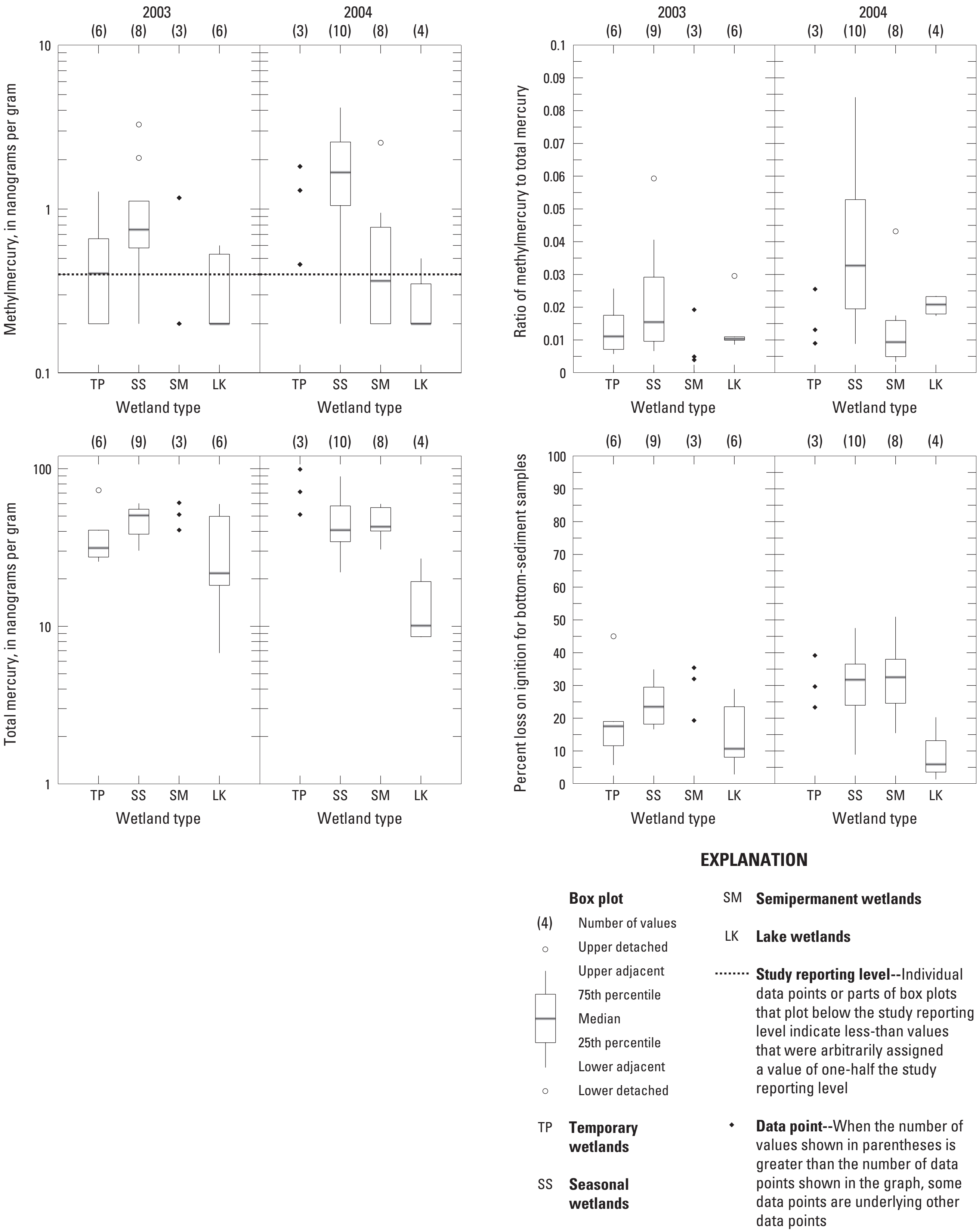


Figure 5. Concentrations of selected mercury constituents in bottom-sediment samples and percent loss on ignition for bottom-sediment samples.

than in 2003, possibly indicating that less inorganic Hg was contributed to the Refuge wetlands in 2004 than in 2003. Differences in climatic conditions and runoff to the wetlands between years might have resulted in smaller contributions of new Hg to the Refuge wetlands in 2004 than in 2003.

Whole-water THg concentrations ranged from 0.87 ng/L for site 3 (SM1THL04) for 2004 to 17.2 ng/L for site 38 (SS1KRU03) for 2003 (supplement 4). Most whole-water THg concentrations (the middle 80 percent) were between 1.60 and 8.71 ng/L (fig. 4). For 2003, the median whole-water THg concentration for lake wetlands (1.58 ng/L) was less than that for temporary wetlands (4.52 ng/L) which was less than that for semipermanent wetlands (6.99 ng/L) which was less than that for seasonal wetlands (7.92 ng/L) (fig. 4). For 2004, the median whole-water THg concentration for lake wetlands (2.37 ng/L) was less than that for semipermanent wetlands (3.95 ng/L) which was less than that for seasonal wetlands (4.52 ng/L) which was less than that for temporary wetlands (6.48 ng/L) (fig. 4). Within types, median whole-water THg concentrations were larger for 2003 than for 2004 for seasonal and semipermanent wetlands and were smaller for 2003 than for 2004 for temporary and lake wetlands. For wetlands sampled in both 2003 and 2004, whole-water THg concentrations were larger for 2003 than for 2004 for site 38 (SS1KRU03) and site 27 (SM1CNC03), were about equal for 2003 and 2004 for site 30 (TP1ASP03) and site 40 (LK1KRU03), and were smaller for 2003 than for 2004 for site 12 (LK1THL03).

The ratio of whole-water MeHg to THg (MeHg:THg) generally provides an index of how efficiently Hg is methylated in aquatic systems. Whole-water MeHg:THg ratios ranged from 0.0006 for site 43 (LK1ELS03) for 2004 to 0.556 for site 38 (SS1KRU03) for 2003 (supplement 4). Most whole-water MeHg:THg ratios (the middle 80 percent) were between 0.026 and 0.317 (fig. 4). For 2003, the median whole-water MeHg:THg ratio for lake wetlands (0.065) was less than that for temporary wetlands (0.122) which was less than that for seasonal wetlands (0.176) which was less than that for semipermanent wetlands (0.289) (fig. 4). For 2004, the median whole-water MeHg:THg ratio for temporary wetlands (0.044) was less than that for seasonal wetlands (0.065) which was less than that for lake wetlands (0.071) which was less than that for semipermanent wetlands (0.123) (fig. 4). Within types, median whole-water MeHg:THg ratios were larger for 2003 than for 2004 except for lake wetlands. For wetlands sampled in both 2003 and 2004, whole-water MeHg:THg ratios were larger for 2003 than for 2004 for site 30 (TP1ASP03), site 38 (SS1KRU03), site 27 (SM1CNC03), and site 12 (LK1THL03) and were smaller for 2003 than for 2004 for site 40 (LK1KRU03).

Bottom-sediment MeHg concentrations ranged from less than 0.4 nanogram per gram (ng/g; dry weight) for several sites for both 2003 and 2004 to 4.16 ng/g for site 24 (SS3CNC04) for 2004 (supplement 4). Most bottom-sediment MeHg concentrations (the middle 80 percent) were between less than 0.4 and 2.16 ng/g (fig. 5). For 2003, the median bottom-sediment MeHg concentration for semipermanent and lake wetlands (less than 0.4 ng/g) was less than that for temporary wetlands (0.41 ng/g) which was less than that for seasonal wetlands (0.75 ng/g) (fig. 5). For 2004, the median bottom-sediment MeHg concentration for semipermanent and lake wetlands (less than 0.4 ng/g) was less than that for temporary wetlands (1.30 ng/g) which was less than that for seasonal wetlands (1.68 ng/g) (fig. 5). Within types, median bottom-sediment MeHg concentrations were smaller for 2003 than for 2004 for temporary and seasonal wetlands and were about equal for 2003 and 2004 for semipermanent and lake wetlands. For wetlands sampled in both 2003 and 2004, bottom-sediment MeHg concentrations were smaller for 2003 than for 2004 for site 30 (TP1ASP03), site 38 (SS1KRU03), and site 27 (SM1CNCO3) and were about equal between 2003 and 2004 for site 40 (LK1KRU03) and site 12 (LK1THL03).

Bottom-sediment THg concentrations ranged from 6.77 ng/g for site 36 (LK1WWH03) for 2003 to 99.0 ng/g for site 7 (TP2WLD04) for 2004 (supplement 4). Most bottom-sediment THg concentrations (the middle 80 percent) were between 19.7 and 62.4 ng/g (fig. 5). For 2003, the median bottom-sediment THg concentration for lake wetlands (21.7 ng/g) was less than that for temporary wetlands (31.4 ng/g) which was less than that for seasonal wetlands (50.5 ng/g) which was less than that for semipermanent wetlands (51.2 ng/g) (fig. 5). For 2004, the median bottom-sediment THg concentration for lake wetlands (10.1 ng/g) was less than that for seasonal wetlands (40.8 ng/g) which was less than that for semipermanent wetlands (42.8 ng/g) which was less than that for temporary wetlands (71.3 ng/g). Within types, median bottom-sediment THg concentrations were smaller for 2003 than for 2004 for temporary wetlands and were larger for 2003 than for 2004 for seasonal, semipermanent, and lake wetlands. For wetlands sampled in both 2003 and 2004, bottom-sediment THg concentrations were larger for 2003 than for 2004 for site 12 (LK1THL03), were about equal for 2003 and 2004 for site 30 (TP1ASP03), site 38 (SS1KRU03), and site 40 (LK1KRU03), and were smaller for 2003 than for 2004 for site 27 (SM1CNCO3).

Bottom-sediment MeHg:THg ratios ranged from 0.003 for site 2 (SM4WLD04) for 2004 to 0.084 for site 38 (SS1KRU03) for 2004. Most bottom-sediment MeHg:THg ratios (the middle 80 percent) were between 0.006 and 0.044 (fig. 5). For 2003, the median bottom-sediment MeHg:THg ratio for semipermanent wetlands (0.005) was less than that for lake wetlands (0.01) which was less than that for temporary wetlands (0.011) which was less than that for seasonal wetlands (0.015) (fig. 5). For 2004, the median bottom-sediment MeHg:THg ratio for semipermanent wetlands (0.009) was less than that for temporary wetlands (0.013) which was less than that for lake wetlands (0.021) which was less than that for seasonal wetlands (0.033) (fig. 5). Within types, median bottom-sediment MeHg:THg ratios were about equal for 2003 and 2004 for temporary wetlands and were smaller for 2003 than for 2004 for seasonal, semipermanent, and lake wetlands. For wetlands sampled in both 2003 and 2004, bottom-sediment MeHg:THg ratios were smaller for 2003 than for 2004 for all sites.

Bottom-sediment percent loss on ignition (LOI) is an index of the amount of organic material in bottom sediment; LOI increases with increasing concentrations of organic material. Bottom-sediment LOI values ranged from 1.3 percent for site 12 (LK1THL03) for 2004 to 51.0 percent for site 19 (SM2CNC04) for 2004 (supplement 4). For 2003, the median bottom-sediment LOI value for lake wetlands (10.7 percent) was less than that for temporary wetlands (17.6 percent) which was less than that for seasonal wetlands (23.5 percent) which was less than that for semipermanent wetlands (32.0 percent) (fig. 5). For 2004, the median bottom-sediment LOI value for lake wetlands (5.9 percent) was less than that for temporary wetlands (29.6 percent) which was less than that for seasonal wetlands (31.8 percent) which was less than that for semipermanent wetlands (32.5 percent) (fig. 5). Within types, median bottom-sediment LOI values were smaller for 2003 than for 2004 for temporary, seasonal, and lake wetlands and were about equal for 2003 and 2004 for semipermanent wetlands. For wetlands sampled in both 2003 and 2004, bottom-sediment LOI values were larger for 2003 than for 2004 for site 30 (TP1ASP03) and site 12 (LK1THL03) and were smaller for 2003 than for 2004 for site 38 (SS1KRU03), site 27 (SM1CNC03), and site 40 (LK1KRU03).

The Refuge wetlands vary in concentrations of Hg constituents and values of bottom-sediment properties among wetland types and between years. Notable patterns in concentrations of Hg constituents and LOI values are as follow: (1) whole-water MeHg concentrations generally were larger for seasonal and semipermanent wetlands than for temporary and lake wetlands; (2) whole-water MeHg concentrations for all wetland types were substantially larger for 2003 than for 2004; (3) inorganic Hg concentrations generally were larger for temporary, seasonal, and semipermanent wetlands than for lake wetlands; (4) inorganic Hg concentrations for all wetland types generally were similar for 2003 and 2004; (5) whole-water MeHg:THg ratios generally were larger for seasonal and semipermanent wetlands than for temporary and lake wetlands; and (6) whole-water MeHg:THg ratios for all wetland types generally were larger for 2003 than for 2004.

Water-Quality Properties and Constituents Measured at Time of Sample Collection

Statistical summaries of water-quality property and constituent values are shown in figure 6. Values varied among wetland types and between years (fig. 6). Turbidity (an index of suspended material) values ranged from less than 1 formazin nephelometric unit (FNU) for several sites for both 2003 and 2004 to 500 FNU for site 42 (SM1WLL04) for 2004 (supplement 2). Median turbidity values for all wetland types were intermediate (between about 5 and 30 FNU) for both 2003 and 2004, and overlap in the distribution of turbidity generally occurred among wetland types (fig. 6). Within types, median turbidity values were similar for both 2003 and 2004 for temporary wetlands (values generally were less than about 15 FNU) and were larger for 2003 than for 2004 for seasonal, semipermanent, and lake wetlands (fig. 6). For wetlands sampled in both 2003 and 2004, differences in turbidity values between years were variable (supplement 2). Notable patterns in turbidity are as follow: (1) turbidity values for the Refuge wetlands generally were moderate, and (2) differences in turbidity patterns among wetland types were not substantial.

Dissolved-oxygen concentrations ranged from 1.3 milligrams per liter (mg/L) for site 38 (SS1KRU03) for 2003 to 17.5 mg/L for site 15 (SS2WLD04) for 2004 (supplement 2). Diel variability in dissolved-oxygen concentrations within a given wetland can be substantial. Because individual wetlands were sampled at varying times of day, comparisons of dissolved-oxygen concentrations between wetlands and among wetland types might be substantially affected by sampling times. Thus, these comparisons should be considered qualitative and used with caution. Substantial overlap in the distribution of dissolved oxygen generally occurred among wetland types (fig. 6), but concentrations tended to be smallest in seasonal and semipermanent wetlands. Within types, median dissolved-oxygen concentrations generally were smaller for 2003 than for 2004. For wetlands sampled in both 2003 and 2004, dissolved-oxygen concentrations also generally were smaller for 2003 than for 2004 (supplement 2).

Patterns in dissolved-oxygen concentrations in the Refuge wetlands are quite variable. The wetlands are relatively shallow and subject to aeration through wind-generated turbulence. Photosynthesis by wetland vegetation also can contribute dissolved oxygen to the water column, especially in the wetlands where subemergent vegetation is predominant. However, the wetlands also are rich in decaying organic material, and the breakdown of the organic material with associated respiration can consume dissolved oxygen quickly. The processes of aeration, photosynthesis, and respiration interact to produce relatively large variability in dissolved-oxygen concentrations in the wetlands and can have a substantial role in chemical reactions that are dependent on oxidation/reduction conditions.

pH values ranged from 6.5 standard units for site 38 (SS1KRU03) for 2003 to 9.8 standard units for site 43 (LK1ELS04) for 2004 (supplement 2). Although some overlap in the distribution of pH generally occurred among wetland types, an obvious trend exists between the length of hydroperiod and pH. The median pH values for temporary and seasonal wetlands were less than the median pH value for semipermanent wetlands which was less than the median pH value for lake wetlands for both 2003 and 2004 (fig. 6). Within types, pH values generally were smaller for 2003 than for 2004. For wetlands sampled in both 2003 and 2004, pH values also were smaller for 2003 than for 2004 (supplement 2).

The smaller pH values for temporary and seasonal wetlands likely are a result of recent atmospheric origin of the surface water contained in the wetlands. Based on data from three National Atmospheric Deposition Program data-collection sites, average pH in North Dakota precipitation ranged from about 5.5 to about 6.0 standard units during 2003–04 (National Atmospheric Deposition Program, 2007). Factors contributing

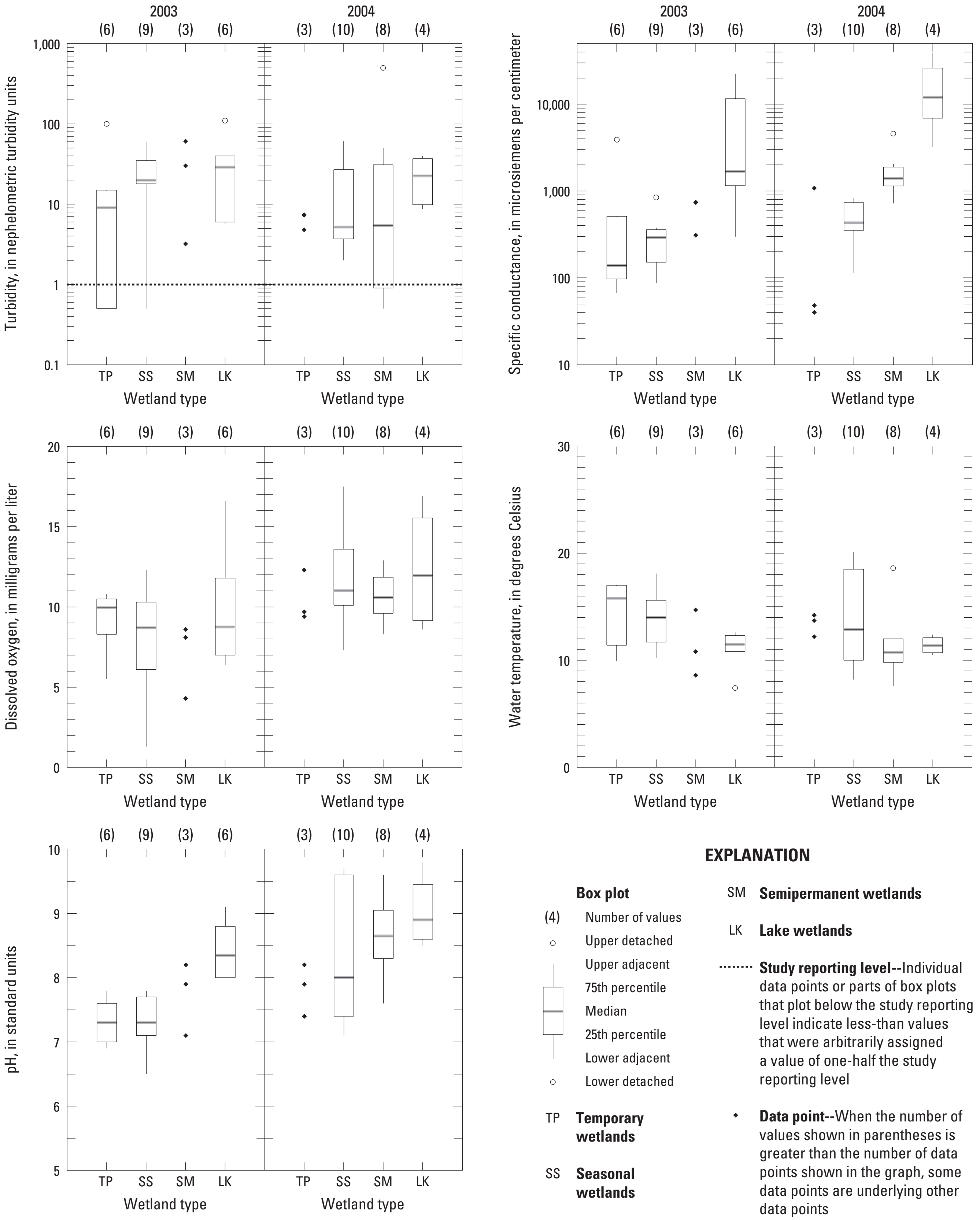


Figure 6. Water-quality property and constituent values measured at time of sample collection.

to the variability in pH in these wetland types include concentrations of organic acids, exchange of dissolved carbon dioxide between the wetland water and the atmosphere, dissolution of carbonate minerals as water interacts with geologic materials, and biological processes (including photosynthesis and respiration) that either consume or produce dissolved carbon dioxide that forms carbonic acid and lowers pH. Sulfate reduction also can affect pH by production of bicarbonate ions and consumption of hydronium ions. For environmental Hg studies, consideration of pH is important because Hg and MeHg frequently are shown to be inversely related to pH (Wiener and others, 2003), and low pH often is associated with high MeHg production rates.

Specific-conductance (an index of dissolved solids) values ranged from 40 microsiemens per centimeter (μS/cm) for site 30 (TP1ASP03) for 2003 to 38,800 μS/cm for site 43 (LK1ELS04) for 2004 (supplement 2). The median specific-conductance value for temporary wetlands was less than that for seasonal wetlands which was less than that for semipermanent wetlands which was less than that for lake wetlands for both 2003 and 2004 (fig. 6). Within types, median specific-conductance values were larger for 2003 than for 2004 for temporary wetlands and were smaller for 2003 than for 2004 for seasonal, semipermanent, and lake wetlands. For wetlands sampled in both 2003 and 2004, specific-conductance values were larger for 2003 than for 2004 for site 30 (TP1ASP03) and were smaller for 2003 than for 2004 for site 38 (SS1KRU03), site 27 (SM1CNC03), site 40 (LK1KRU03), and site 12 (LK1THL03) (supplement 2).

Differences in specific-conductance values among wetland types probably largely reflect hydrogeologic characteristics of the wetlands, the extent to which the wetland waters have interacted with geologic materials, and the pathway of infiltrating water entering the wetland. Temporary and seasonal wetlands predominantly contain water of atmospheric origin that has fallen directly on the wetlands with relatively little opportunity to interact with geologic materials. Thus, these wetland types tend to have smaller specific-conductances values than the other wetland types. Semipermanent and lake wetlands contain water in part contributed by ground water that has had prolonged contact with geologic materials. Thus, these wetland types tend to have larger specific-conductances values than the other wetland types. Also, because water remains in the semipermanent and lake wetlands for greater durations than in the other wetlands, more opportunity exists for evaporative concentration of dissolved salts.

Water temperatures ranged from 7.4°C for site 12 (LK1THL03) for 2003 to 20.1°C for site 15 (SS2WLD04) for 2004 (supplement 2). Diel variability in water temperature within a given wetland can be substantial. Because individual wetlands were sampled at varying times of day, comparisons of water temperature between wetlands and among wetland types might be substantially affected by sampling times. Thus, these comparisons should be considered qualitative and used with caution. Although overlap in the distribution of water temperatures occurred among wetland types, the median water temperature for temporary wetlands was higher than that for seasonal wetlands which was higher than those for semipermanent and lake wetlands for both 2003 and 2004 (fig. 6). Within types, water temperatures were slightly higher for 2003 than for 2004 for temporary and seasonal wetlands and were about equal for 2003 and 2004 for semipermanent and lake wetlands. For wetlands sampled in both 2003 and 2004, water temperatures were about equal for 2003 and 2004, varying by less than about 3°C (supplement 2).

Variability in water temperatures among wetland types probably reflects geomorphologic characteristics of the wetlands. Temporary and seasonal wetlands are shallower than the other wetlands, and water temperatures increase more quickly with rising air temperatures in the spring. Semipermanent and lake wetlands are deeper than the other wetlands and have greater volumes, and water temperatures respond more slowly to rising air temperatures in the spring. Differences in water temperatures between 2003 and 2004 appear to be relatively small. Water temperature can have a substantial role in Hg environmental chemistry because high water temperatures tend to increase bacterial metabolic rates, which can result in high rates of sulfate reduction and increase Hg methylation.

Notable patterns in water-quality properties and constituents are as follow: (1) overlap in the distribution of dissolved oxygen generally occurred among wetland types, but concentrations tended to be smallest in seasonal and semipermanent wetlands and generally were smaller for 2003 than for 2004 for all wetland types; (2) pH values tended to be smaller for temporary and seasonal wetlands than for semipermanent and lake wetlands and were substantially smaller for 2003 than for 2004 for all wetland types; and (3) specific-conductance values generally were smaller for temporary wetlands than for the other wetlands, were intermediate for seasonal and semipermanent wetlands, were larger for lake wetlands than for the other wetlands, and were substantially smaller for 2003 than for 2004 for all wetland types except temporary wetlands.

Major-Ion and Selected Trace-Element Constituents in Water Samples

Statistical summaries of concentrations of dissolved solids and dissolved sulfate are shown in figure 7. Proportions of major ions for wetlands that were sampled in both 2003 and 2004 are shown in figure 8. The average percent difference between milliequivalent concentrations of cations and anions was 2.6 percent for all samples collected during this study. The greatest deviation from zero for the percent difference between cations and anions for any sample was -8.4 percent. About 90 percent of all samples had absolute values of percent differences between cations and anions that were less than 5 percent. These percent differences between milliequivalent concentrations of cations and anions indicate overall acceptable performance of the sample-collection and -analysis process and also indicate no constituents that substantially contribute to the charge balance were omitted.

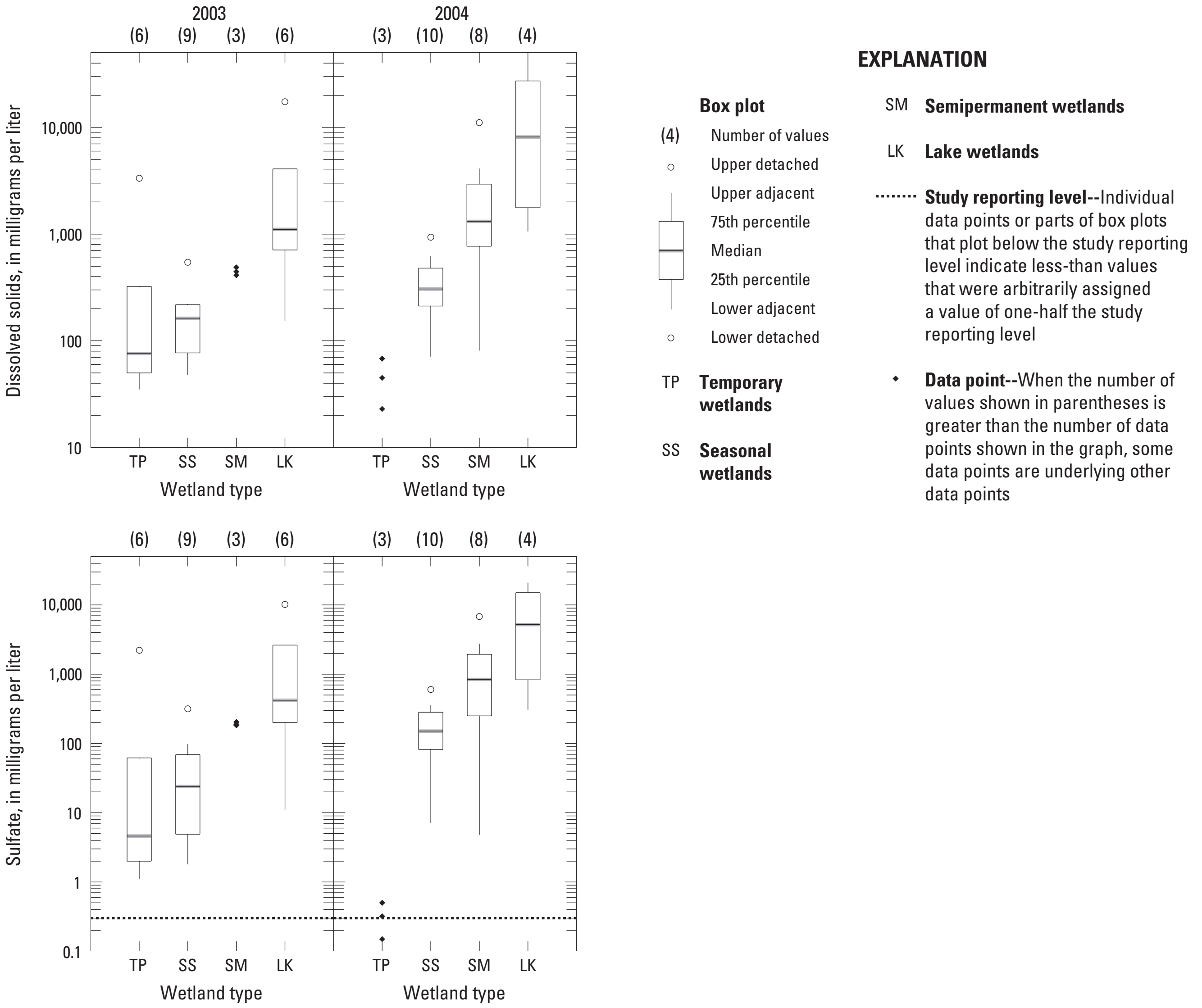


Figure 7. Concentrations of dissolved solids and dissolved sulfate in water samples.

Major-ion relations in prairie-pothole wetlands often are complex, largely because of variability in surface-water/ground-water interactions and frequency and duration of inundation. LaBaugh (1989) presented a detailed discussion on major-ion chemistry and interactions in prairie pothole wetlands. Although a detailed analysis and discussion of major-ion chemistry in the Refuge wetlands is outside the scope of this report, sulfate is of particular importance to Hg processing in aquatic environments and warrants discussion.

Dissolved sulfate is particularly relevant to Hg environmental chemistry because inorganic Hg is converted to MeHg as a byproduct of bacterially mediated sulfate reduction. Dissolved-sulfate concentrations generally must exceed about 1 mg/L to sustain sulfate reduction in wetlands (Gilmour and Krabbenhoft, 2001); however, if sulfate concentrations are too large, the accumulation of free sulfide (a product of sulfate reduction) in pore waters can have an inhibitory effect on the MeHg formation process (U.S. Department of Commerce, National Oceanic and Atmospheric Administration, National Oceanic Service, 1996; Benoit and others, 1999; Benoit and others, 2001). Factors that contribute to sulfide formation and accumulation can be complex and are not necessarily restricted to variability in sulfate-reduction rates. Thus, a complex, non-linear relation between sulfate concentration and MeHg exists for aquatic ecosystems (Wiener and others, 2003).

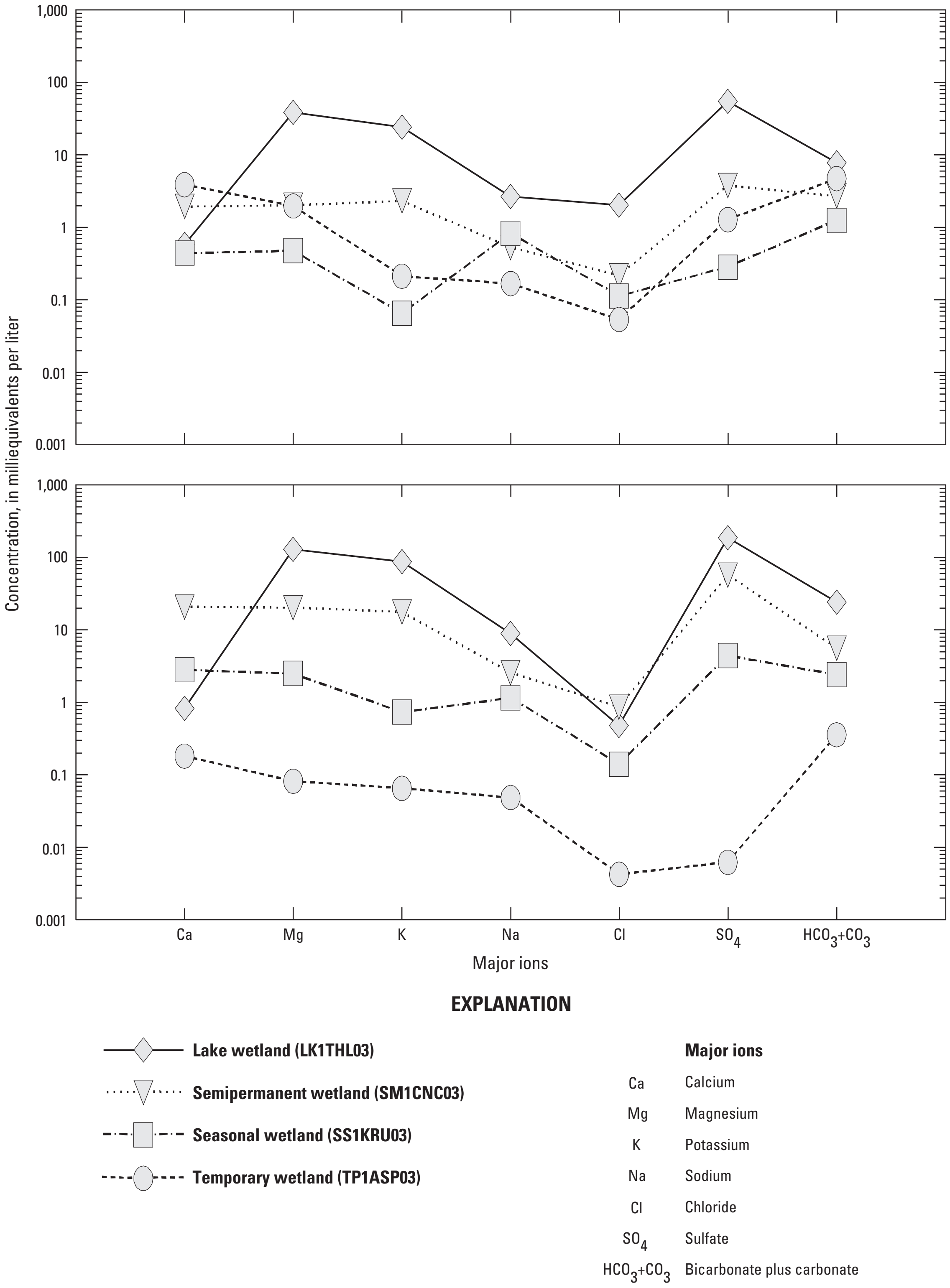


Figure 8. Concentrations and proportions of major ions for wetlands sampled in both 2003 and 2004 (diagrams are modified from Schoeller, 1959).

Dissolved-sulfate concentrations ranged from less than 0.30 mg/L for site 7 (TP2WLD04) for 2004 to 21,000 mg/L for site 43 (LK1ELS04) for 2004 (supplement 3). The median dissolved-sulfate concentrations for temporary wetlands (4.6 and 0.32 mg/L for 2003 and 2004, respectively) were less than those for seasonal wetlands (24 and 152 mg/L for 2003 and 2004, respectively) which were less than those for semipermanent wetlands (189 and 841 mg/L for 2003 and 2004, respectively) which were less than those for lake wetlands (422 and 5,200 mg/L for 2003 and 2004, respectively) (fig. 7). Within types, dissolved-sulfate concentrations generally were larger for 2003 than for 2004 for temporary wetlands and were smaller for 2003 than for 2004 for seasonal, semipermanent, and lake wetlands. For wetlands sampled in both 2003 and 2004, dissolved-sulfate concentrations were larger for 2003 than for 2004 for site 30 (TP1ASP03) and were smaller for 2003 than for 2004 for site 38 (SS1KRU03), site 27 (SM1CNC03), site 40 (LK1KRU03), and site 12 (LK1THL03). About 90 percent of the wetlands had dissolved-sulfate concentrations that exceeded 2 mg/L, which is near the concentration below which substantial sulfate reduction generally does not occur. However, only about 50 percent of the temporary wetlands had dissolved-sulfate concentrations that exceeded 2 mg/L. Adequate dissolved sulfate might be a factor that limits Hg methylation in temporary wetlands in the Refuge. Sulfate concentrations in lake wetlands generally are large, and the accumulation of large concentrations of free sulfide in pore waters of the lake wetlands might be limiting Hg methylation in those settings.

Patterns in major-ion concentrations and proportions among wetland types and between years are shown in figure 8 for selected wetlands that were sampled in both 2003 and 2004. Proportions of major ions vary among wetland types and between years (fig. 8). Sulfate generally accounts for a smaller proportion of major ions in temporary and seasonal wetlands than in semipermanent and lake wetlands (fig. 8). Two factors probably contribute to the differences in sulfate proportions among wetland types. These factors are as follow: (1) temporary and seasonal wetlands generally contain water of recent atmospheric origin that has had relatively little opportunity to interact with geologic materials, and (2) semipermanent and lake wetlands receive a greater contribution of water from ground water. The proportion of sulfate among major ions typically increases as water moves through geologic materials (LaBaugh, 1989). This probably contributes to a larger proportion of sulfate among major ions in semipermanent and lake wetlands than in temporary and seasonal wetlands. The proportion of sulfate among major ions also typically increases during evaporative concentration and accompanying mineral precipitation processes (Eugster and Jones, 1979). Semipermanent and lake wetlands generally are inundated for longer periods of time than temporary and seasonal wetlands. Thus, effects of evaporative concentration are more pronounced in semipermanent and lake wetlands than in temporary and seasonal wetlands, further increasing the sulfate proportions among major ions. The proportion of sulfate among major ions was larger in 2004 than in 2003 for all wetland types except temporary wetlands (fig. 8). Runoff to the Refuge wetlands in 2004 probably was substantially smaller than in 2003 and resulted in smaller contributions of dilute atmospheric water in 2004. The decrease in dilution in 2004 might have increased effects of evaporative concentration and mineral precipitation processes, resulting in greater proportions of sulfate for most wetland types in 2004.

Patterns in dissolved-solids concentrations among wetland types and between years (fig. 7) are similar to those for specific conductance (fig. 6), which is an index of dissolved-solids concentration. The patterns for specific conductance and the factors that contribute to the patterns were discussed previously in this report.

Notable patterns in major-ion constituents for the Refuge wetlands are as follow: (1) dissolved-solids concentrations were smaller for temporary wetlands than for the other wetlands, were intermediate for seasonal and semipermanent wetlands, and were larger for lake wetlands than for the other wetlands for both 2003 and 2004; (2) dissolved-solids concentrations were substantially smaller for 2003 than for 2004 for all wetland types except temporary wetlands; (3) dissolved-sulfate concentrations varied among wetland types and between years in a manner similar to that for dissolved-solids concentrations; (4) the proportion of sulfate among major ions was larger for semipermanent and lake wetlands than for temporary and seasonal wetlands and also for 2004 than for 2003 for all wetland types except temporary wetlands; (5) sulfate concentrations in temporary wetlands generally were small and might limit sulfate reduction; and (6) sulfate concentrations in lake wetlands generally were large, and the accumulation of large concentrations of free sulfide might limit availability of inorganic Hg for methylation.

Although water samples were analyzed for many non-Hg trace elements (table 4), statistical summaries of concentrations were evaluated only for barium and manganese (fig. 9). Preliminary exploratory data analyses, which consisted of plotting non-Hg trace-element concentrations in relation to Hg-constituent concentrations and computing Pearson correlation coefficients for non-Hg trace elements and Hg constituents, were conducted for all non-Hg trace elements. The specific results of these preliminary exploratory data analyses are not presented in this report. However, the results of these analyses indicated that most trace elements either have concentrations that are not significantly correlated with concentrations of Hg constituents, have too few values that are greater than study reporting levels to warrant detailed discussion, or do not have substantial relevance to Hg processes. Discussion of analytical results for non-Hg trace elements is restricted to those trace elements for which concentrations are significantly correlated with concentrations of one or more Hg constituents, have more than about 25 percent of values greater than study reporting levels, and might have potential relevance to Hg processes.

Dissolved-barium concentrations ranged from less than 1.0 µg/L for site 36 (LK1WWH03) for 2003 to 226 µg/L for site 8 (SM2WLD04) for 2004 (supplement 3). For both 2003 and 2004, the median dissolved-barium concentrations for temporary and lake wetlands were less than those for seasonal and

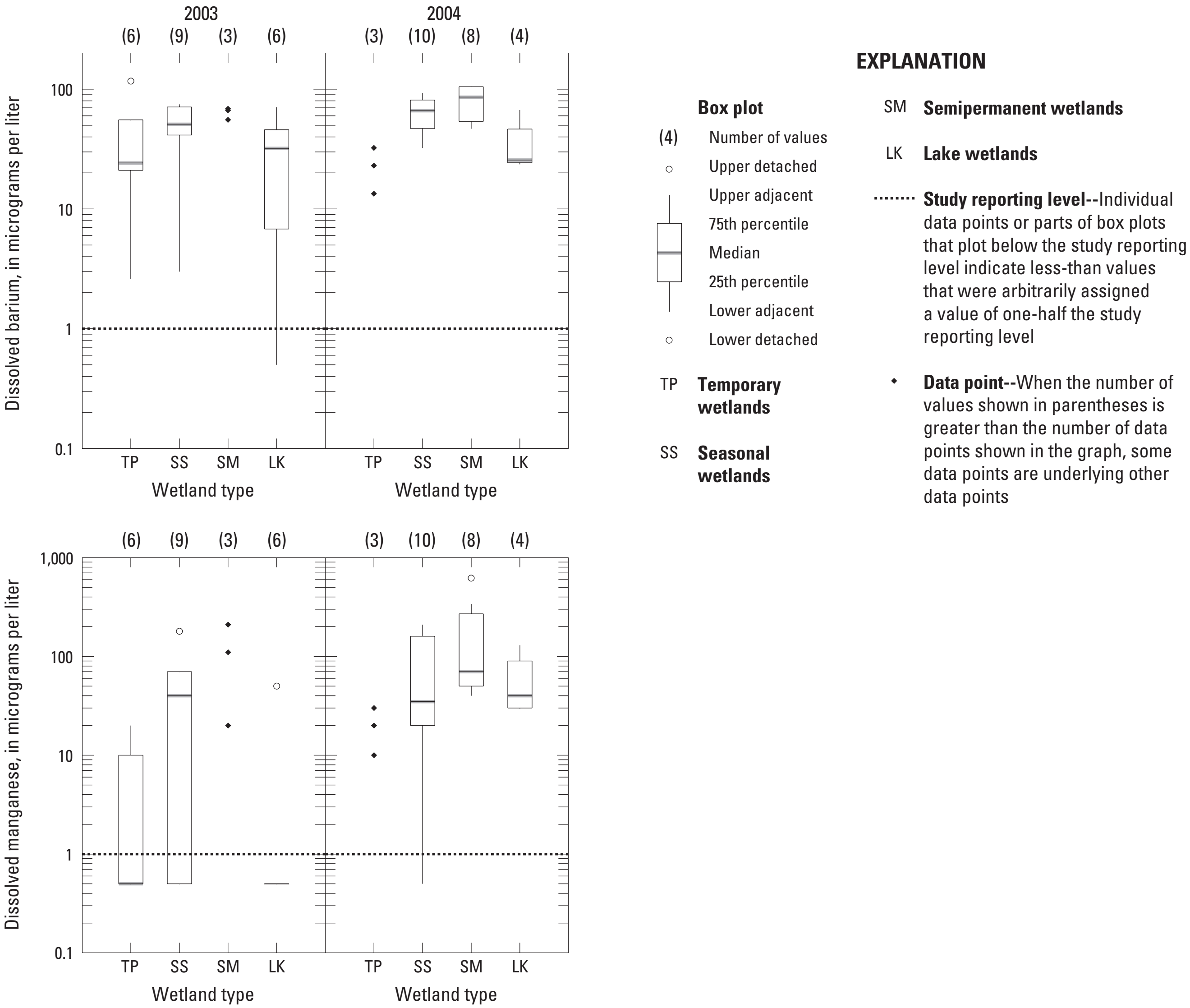


Figure 9. Concentrations of selected nonmercury trace elements in water samples.

semipermanent wetlands (fig. 9). Within types, median dissolved-barium concentrations were about equal for 2003 and 2004 for temporary and lake wetlands and were smaller for 2003 than for 2004 for seasonal and semipermanent wetlands. For wetlands sampled in both 2003 and 2004, dissolved-barium concentrations were larger for 2003 than for 2004 for site 30 (TP1ASP03) and site 38 (SS1KRU03) and were smaller for 2003 than for 2004 for site 27 (SM1CNC03), site 40 (LK1KRU03), and site 12 (LK1THL03). Barium solubility is strongly linked to oxidation-reduction processes, especially those involving adsorption/desorption from oxide and hydroxide compounds (Hem, 1985). Barium solubility is favored in less oxidized conditions. Therefore, patterns in barium concentrations might provide information on variability in oxidation-reduction conditions at the sediment/water interface in the Refuge wetlands.

Dissolved-manganese concentrations ranged from less than 10 µg/L for several sites for both 2003 and 2004 to 620 µg/L for site 27 (SM1CNC03) for 2004 (supplement 3). For 2003, the median dissolved-manganese concentrations for temporary and lake wetlands were less than the study reporting level and were less than those for seasonal and semipermanent wetlands (40 and 110 µg/L, respectively) (fig. 9). For 2004, the median dissolved-manganese concentration for temporary wetlands (20 µg/L) was less than those for seasonal, semipermanent, and lake wetlands (35, 70, and 40 µg/L, respectively).

Within types, median dissolved-manganese concentrations were smaller for 2003 than for 2004 for temporary and lake wetlands, were equal for 2003 and 2004 for seasonal wetlands, and were larger for 2003 than for 2004 for semipermanent wetlands. For wetlands sampled in both 2003 and 2004, dissolved-manganese concentrations were larger for 2003 than for 2004 for site 30 (TP1ASP03) and were smaller for 2003 than for 2004 for site 38 (SS1KRU03), site 27 (SM1CNC03), site 40 (LK1KRU03), and site 12 (LK1THL03). As for barium, manganese solubility is strongly linked to oxidation-reduction processes, especially those involving adsorption/desorption from clays and oxide and hydroxide compounds (Hem, 1985). Oxidation-reduction processes that promote solubility of manganese previously have been noted to be associated with production of MeHg (Bonzongo and others, 1996). Patterns in manganese concentrations might provide information on variability in oxidation-reduction conditions at the sediment/water interface in the Refuge wetlands.

The notable pattern in non-Hg trace-element constituents for the Refuge wetlands is that barium and manganese concentrations, which are strongly dependent on oxidation-reduction processes, generally were largest for seasonal and semipermanent wetlands.

Organic-Carbon Constituents in Water Samples

Statistical summaries of concentrations of dissolved and whole-water organic-carbon constituents are shown in figure 10. Nearly all of the organic carbon in surface water of the wetlands was in dissolved phase. For 75 percent of the water samples, DOC accounted for 98 percent or more of whole-water organic carbon, and the smallest proportion of DOC to whole-water organic carbon was 80 percent. Thus, although analytical results for both dissolved and whole-water organic carbon are given in the tables and figures in this report, discussion is limited to only DOC.

DOC is critical to processing of Hg in aquatic systems. DOC forms strong dissolved complexes with both inorganic Hg and MeHg (Stumm and Morgan, 1996), which can maintain Hg in the water column where it is available for various types of biotic and abiotic processing. The ability of DOC to regulate aqueous Hg concentrations is affected by both the amount and composition of DOC (Haitzer and others, 2002 and 2003). DOC largely is derived as a byproduct from microbial processes (including sulfate reduction, the process by which most inorganic Hg in aquatic systems is methylated) that use particulate organic matter in soil and sediments. Large DOC concentrations often are reflective of aquatic systems that have high organic carbon use in bottom sediments. Thus, MeHg and DOC commonly are positively correlated (Krabbenhoft and others, 1999).

DOC concentrations generally are larger in wetlands than in other freshwater aquatic systems (Thurman, 1985). Vegetation in and/or around wetlands tends to be more dense than for many other freshwater aquatic systems and wetlands are accumulation sites for particulate organic matter. Among different kinds of wetlands, DOC concentrations in northern prairie wetlands tend to be high; concentrations routinely exceed 20 mg/L and sometimes exceed 100 mg/L (Arts and others, 2000). Wetland complexes generally have a larger ratio of littoral (that is, in or near the land/water interface) area to open-water area than most other freshwater aquatic systems. The littoral zone supports emergent and submergent aquatic vegetation (with associated epiphytic algae) and accounts for extremely high rates of primary productivity (Wetzel, 1992). When vegetation in or near wetlands dies in the fall (or as wetland water levels recede during summer), the dead particulate organic matter generally is deposited near the production site where it undergoes decomposition, producing soluble organic byproducts. In spring, surface runoff, interflow (that is, water that infiltrates the soil surface, travels generally laterally through the vadose zone, and then discharges to the surface at a point down slope), or interaction between rising water levels and previously dry wetland soils mobilizes soluble organic compounds in various stages of decomposition (Wetzel, 1992) and transports them to the wetland water. Thus, most organic carbon entering prairie wetland water is a complex mixture of dissolved compounds that typically is composed largely of humic substances with high bio- and photoreactivity (Curtis and Adams, 1995; Amon and Benner, 1996; Waiser and Robarts, 2004).

As DOC resides in wetland water, it is progressively degraded by both biotic processes (Tulonen and others, 1992; Amon and Benner, 1996; del Giorgio and Cole, 1998) and abiotic processes (primarily photolysis; Miller, 1998; Waiser and Robarts, 2004). These processes alter the composition of the DOC from higher to lower states of bio- and photoreactivity and also can result in removal of DOC from the water by incorporation into biomass or mineralization to inorganic carbon. Also, high-molecular-weight aromatic compounds that are common in recently derived humic DOC readily adsorb to inorganic particulate materials (Chin and others, 1998; Tarchitzky and Chen, 2002). This process also can result in removal of recently introduced DOC from wetland water. In sum, with increased residence time in a wetland, biotic and abiotic processes tend to result in alteration of DOC from higher to lower states of bio- and photoreactivity and also reduce the mass of DOC in the water.

In semiarid regions, where evaporation exceeds precipitation on a mean annual basis, evaporative concentration can affect DOC concentrations. As DOC ages in wetland water, it can eventually reach a state where it is very resistant to further biotic or abiotic degradation, is generally conservative, and is subject to evaporative concentration (Curtis and Adams, 1995; Waiser and Robarts, 2004).

Characteristics of DOC processing in prairie pothole wetlands have important implications for the different wetland types in the Refuge. Temporary and seasonal wetlands typically are not inundated throughout the year in most years (table 1; Cowardin and others, 1979). DOC in these wetlands occurs largely as a result of rising water levels flooding previously dry wetland soils, mobilizing soluble organic compounds, and stimulating organic carbon use and DOC production. DOC also can be contributed to these wetlands by surface runoff and interflow. Because of shorter inundation times in the temporary and seasonal wetlands than in the other wetlands, less opportunity exists for biotic and abiotic processing of organic carbon in the wetland water column. Semipermanent wetlands are inundated throughout the year in some years, and lake wetlands are inundated throughout the year in nearly all years (Cowardin and others, 1979). These wetlands receive new DOC during spring melt periods and runoff events. However, organic carbon can remain in the wetland water column for relatively long periods in semipermanent wetlands and for very long periods in lake wetlands and can undergo extensive biotic and abiotic processing that can affect both the concentration and composition of DOC (Tulonen and others, 1992; Amon and Benner, 1996; del Giorgio and Cole, 1998; Miller, 1998; Waiser and Robarts, 2004). Thus, DOC in temporary and seasonal wetlands might be expected to consist of a larger proportion of reactive DOC than DOC in semipermanent and lake wetlands. Another factor that might contribute to differences in DOC concentrations among wetland types is that seasonal and semipermanent wetlands generally have more dense vegetation growth in and near the wetlands, which might result in greater contributions of new DOC to these wetland types during runoff periods.

DOC concentrations ranged from 8.2 mg/L for site 30 (TP1ASP03) for 2004 to 154 mg/L for site 43 (LK1ELS04) for 2004 (supplement 4). For 2003, the median DOC concentrations for temporary and lake wetlands (22.4 and 24.5 mg/L,

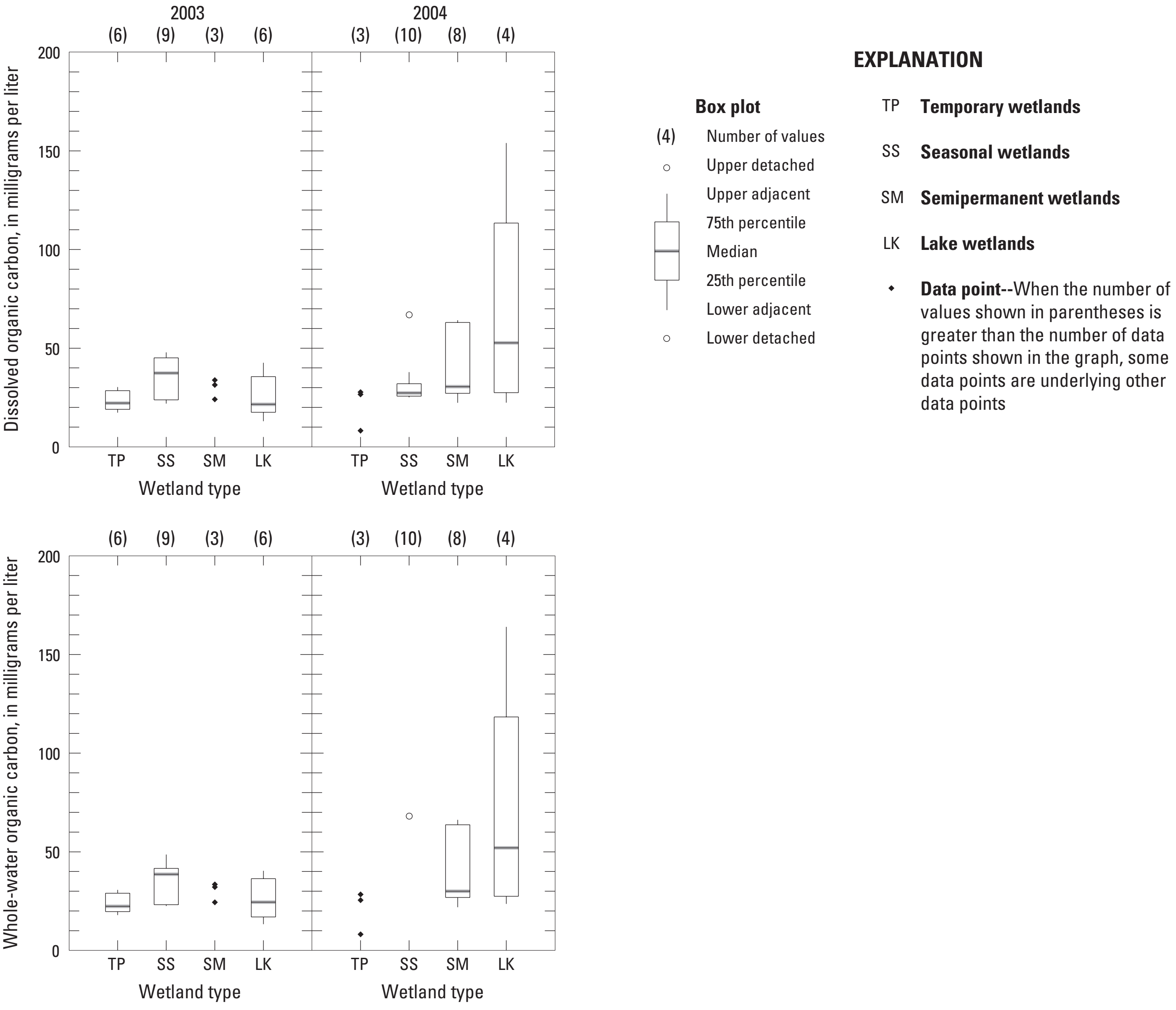


Figure 10. Concentrations of organic-carbon constituents in water samples.

respectively) were less than those for seasonal and semipermanent wetlands (38.6 and 32.4 mg/L, respectively) (fig. 10). These DOC concentrations are large relative to those in most natural aquatic systems, probably demonstrating high levels of carbon use in the sediments of the Refuge wetlands. For 2004, the median DOC concentrations for temporary, seasonal, and semipermanent wetlands were similar (25.5, 28.0, and 30.0 mg/L, respectively) and were less than the median for lake wetlands (52.0 mg/L). Within types, median DOC concentrations generally were similar for 2003 and 2004 for temporary and semipermanent wetlands, were larger for 2003 than for 2004 for seasonal wetlands, and were smaller for 2003 than for 2004 for lake wetlands. For wetlands sampled in both 2003 and 2004, DOC concentrations were larger in 2003 than in 2004 for site 30 (TP1ASP03) and were smaller for 2003 than for 2004 for site 38 (SS1KRU03), site 27 (SM1CNC03), site 40 (LK1KRU03), and site 12 (LK1THL03). Temporary wetlands generally had smaller DOC concentrations than the other wetland types. For seasonal, semipermanent, and lake wetlands, DOC concentrations generally were relatively large. However, for these types, ranges within types and differences among types and between years generally were quite variable.

Notable patterns in DOC concentrations are as follow: (1) during the wet climatic conditions of 2003, DOC concentrations for seasonal and semipermanent wetlands were larger than those for temporary and lake wetlands; (2) DOC concentrations for seasonal wetlands generally were substantially larger for 2003 than 2004, and (3) many DOC concentrations for semipermanent and lake wetlands were large for 2004. The wet climatic conditions prior to sample collection in 2003 probably resulted in substantial runoff to the wetlands, substantial flooding of the vegetated wetland margins, and interflow of water through the terrestrial root zone and into the wetlands. These processes could have resulted in relatively large contributions of organic carbon to the wetlands. The dry climatic conditions prior to sample collection in 2004 probably resulted in substantially less runoff into the wetlands than in 2003, smaller inundation areas than in 2003, and less flooding of the vegetated wetland margins than in 2003. Contributions of new energy-rich organic carbon to the Refuge wetlands probably was generally smaller in 2004 than in 2003. The large DOC concentrations in semipermanent and especially lake wetlands in 2004 might have resulted from evaporative concentration of energy-poor DOC that was carried over in the wetlands from the previous fall.

Relations Between Mercury Constituents and Other Properties and Constituents in Water

The primary focus of this study is to document the occurrence of Hg and MeHg in the Refuge wetlands and to investigate possible factors or processes that govern spatial and temporal variability in Hg and MeHg. Relations between concentrations of Hg, MeHg, and other properties and constituents that have been shown to affect Hg cycling in aquatic systems are discussed in this section. Because of the complexity of Hg chemistry in aquatic systems and the limited scope of this study, several assumptions were used: (1) atmospheric deposition of inorganic Hg was relatively spatially uniform throughout the Refuge during the study period; (2) differences in concentrations of MeHg among wetland types and between years primarily represent differences in concentrations of bioavailable inorganic Hg for methylation and/or differences in conditions that affect rates of sulfate reduction and, thus, rates of Hg methylation in the wetlands; (3) differences in concentrations of MeHg among wetland types and between years probably were not strongly affected by differences in rates of demethylation; and (4) differences in concentrations of Hg among wetland types and between years probably was not strongly affected by differences in rates of volatilization of dissolved gaseous Hg. Although specific data to support these assumptions were not collected during this study, the assumptions probably are reasonable. Photochemical reactions can strongly affect concentrations of MeHg through photo-demethylation reactions (Krabbenhoft and others, 2002) and, to a lesser degree, concentrations of inorganic Hg through photoreduction of Hg^{+2} to Hg^0, which can volatilize to the atmosphere (Krabbenhoft and others, 1998). All of the Refuge wetlands are relatively shallow, well mixed as a result of wind-generated turbulence, and elevated in DOC. Previous studies have indicated that ultraviolet-light wavelengths are most responsible for facilitating photochemical reactions that involve Hg and MeHg in surface waters (Krabbenhoft and others, 1998; Krabbenhoft and others, 2002). DOC effectively limits the penetration of ultraviolet light at elevated concentrations. Given that all of the wetlands sampled for this study have relatively large concentrations of DOC, differences in rates of these photochemical processes among wetland types probably do not have important roles in controlling differences in Hg and MeHg concentrations among the wetland types.

Hg methylation in aquatic systems largely occurs as a byproduct of bacterially mediated sulfate reduction, a form of anaerobic respiration (U.S. Department of Commerce, National Oceanic and Atmospheric Administration, National Oceanic Service, 1996; Ullrich and others, 2001). Sulfate reduction requires adequate organic carbon, anaerobic conditions at the water-sediment interface, and adequate sulfate. For MeHg to result from the sulfate-reduction process, dissolved inorganic Hg must be present in a bioavailable form. Although inorganic Hg bioavailability in aquatic systems is not well understood and is presently an active area of research (Wiener and others, 2003), researchers have indicated that (1) DOC, which can complex with Hg and maintain it at elevated concentrations in the water column and pore waters where it is available for biotic uptake and methylation; (2) sulfate, which at small concentrations can be limiting or at large concentrations can result in the accumulation of sulfide in pore waters and have an inhibitory effect on methylation; and (3) other factors, such as Hg loading rates and organic carbon availability, which affect the concentration of bioavailable Hg and microbial process rates, can affect Hg bioavailability.

The relations between MeHg, THg, pH, sulfate, and DOC are complex and difficult to consider independently. Therefore, the relations are illustrated by scatter plots of dissolved solids, DOC, MeHg, and THg concentrations in relation to pH for the various types of Refuge wetlands for 2003 and 2004 combined (fig. 11). LOWESS smooth lines (Cleveland and McGill, 1984; Cleveland, 1985) that show the central tendencies of the relations were used in the scatter plots to help discern the relations among the variables (Helsel and Hirsch, 1992). The smooth lines do not imply statistically significant relations. Rather, because many of the biogeochemical factors that affect the occurrence of Hg and MeHg in natural aquatic systems (including DOC processing, sulfate reduction, and Hg solubility) also affect or are affected by pH, the relatively large pH gradient that occurs across the different wetland types on the Refuge and between the 2 years of sampling provides a convenient index for examining factors that affect Hg and MeHg concentrations in the wetlands. The Refuge wetlands show a clear gradation in pH that is related to the specific wetland's hydroperiod, such that more ephemeral wetlands generally have relatively low pH, and perennially flooded lakes generally have relatively high pH (fig. 6). Presenting relations between dissolved solids, DOC, MeHg, THg, and pH for all wetland types and both 2003 and 2004 data combined might result in an oversimplification of specific processes that could have been occurring in individual wetlands. However, some patterns that probably represent important biogeochemical processes that were likely to have occurred are evident.

In the Refuge wetlands, relatively small dissolved-solids concentrations (less than about 500 mg/L) generally were associated with relatively low pH (values of less than about 8 standard units) (fig. 11*A*) and typically occurred in temporary and seasonal wetlands (although these conditions also occur in some semipermanent wetlands). Relatively large dissolved-solids concentrations generally were associated with relatively high pH and typically occurred in semipermanent and lake wetlands. The relation between dissolved-solids concentration and pH for the Refuge wetlands was moderately strong (Pearson correlation coefficient 0.44) but highly significant (p-value equals 0.001; alpha equals 0.05).

Dissolved-solids concentrations generally increase with increased contact with geologic materials (LaBaugh, 1989). Also, in closed-basin lakes and wetlands, evaporative concentration increases dissolved-solids concentrations with increasing water residence time (Eugster and Jones, 1979). Thus, in closed-basin lakes and wetlands, dissolved-solids concentrations generally increase with residence time (Curtis and Prepas, 1993; Curtis and Adams, 1995). The generally positive relation between dissolved-solids concentration and pH suggests that pH in closed prairie pothole wetlands also generally increases with residence time in the water body. This pattern of relatively large dissolved-solids concentrations being associated with relatively high pH also has been reported by Bierhuizen and Prepas (1985) and Curtis and Adams (1995). However, factors that affect both dissolved-solids concentrations and pH in prairie pothole wetlands are complex and contribute to variability in the general relation between dissolved-solids concentrations and pH.

For the relatively low pH (values of less than about 8 standard units) generally typical of the temporary and seasonal wetlands, relatively large variability occurred in the relation between dissolved solids and pH. In this pH range, the primary determinants of dissolved solids and pH probably are less strongly associated. Several seasonal wetlands and a semipermanent wetland that were sampled in 2004 had relatively high pH (values ranging from 9.5 to 9.7 standard units) and relatively small dissolved-solids concentrations (ranging from 251 to 935 mg/L). This group of wetlands deviates from the general relation between dissolved solids and pH among the Refuge wetlands. A possible explanation for the dissolved-solids and pH characteristics of this group of wetlands is that the water that occurred in the wetlands in 2004 was predominantly carry-over water from 2003 as opposed to new water contributed from runoff or recent precipitation. These conditions might be a relatively infrequent occurrence for these wetlands. Seasonal wetland sampling site SS1KRU03 (specifically labeled in figure 11), which was sampled in both 2003 and 2004 and is among the group of wetlands that deviates from the general relation, provides evidence for the possible explanation. During the wet climatic conditions of 2003, SS1KRU03 had a relatively small dissolved-solids concentration of 108 mg/L and a relatively low pH of 6.5 standard units (fig. 11*A*), indicating the water in the wetland was predominantly of recent atmospheric origin. During the dry climatic conditions of 2004, SS1KRU03 had a dissolved-solids concentration of 442 mg/L and a pH of 9.7 standard units. The relative increase in dissolved-solids concentration between 2003 and 2004 for SS1KRU03 (about fourfold) generally was similar to the increase for a semipermanent and two lake wetlands that were sampled in both 2003 and 2004, were known to have carried over water between years, and were affected by evaporative concentration. If the occurrence of substantial amounts of carry-over water was common for SS1KRU03, the dissolved-solids concentration likely would have substantially increased with time as a result of evaporative concentration, and the relatively small dissolved-solids concentration (108 mg/L) in 2003 would have been expected to be much larger.

Although some exceptions exist, the general relation between dissolved solids and pH in the Refuge wetlands is that relatively small dissolved-solids concentrations are associated with relatively low pH in short hydroperiod wetlands (that is, temporary and seasonal wetlands) and relatively large dissolved-solids concentrations are associated with relatively high pH in long hydroperiod wetlands (that is, semipermanent and permanent wetlands). This general relation is substantially affected by contributions of DOC to and processing of DOC in the Refuge wetlands.

DOC shows a concave (U-shaped) pattern with respect to pH (fig. 11). Relatively large DOC concentrations are associated with both relatively low and relatively high pH, and relatively small DOC concentrations are associated with intermediate pH. The following discussion presents possible explanations for this pattern. Typically, the prairie pothole region dries during summer and fall, resulting in dessication of wetlands soils.

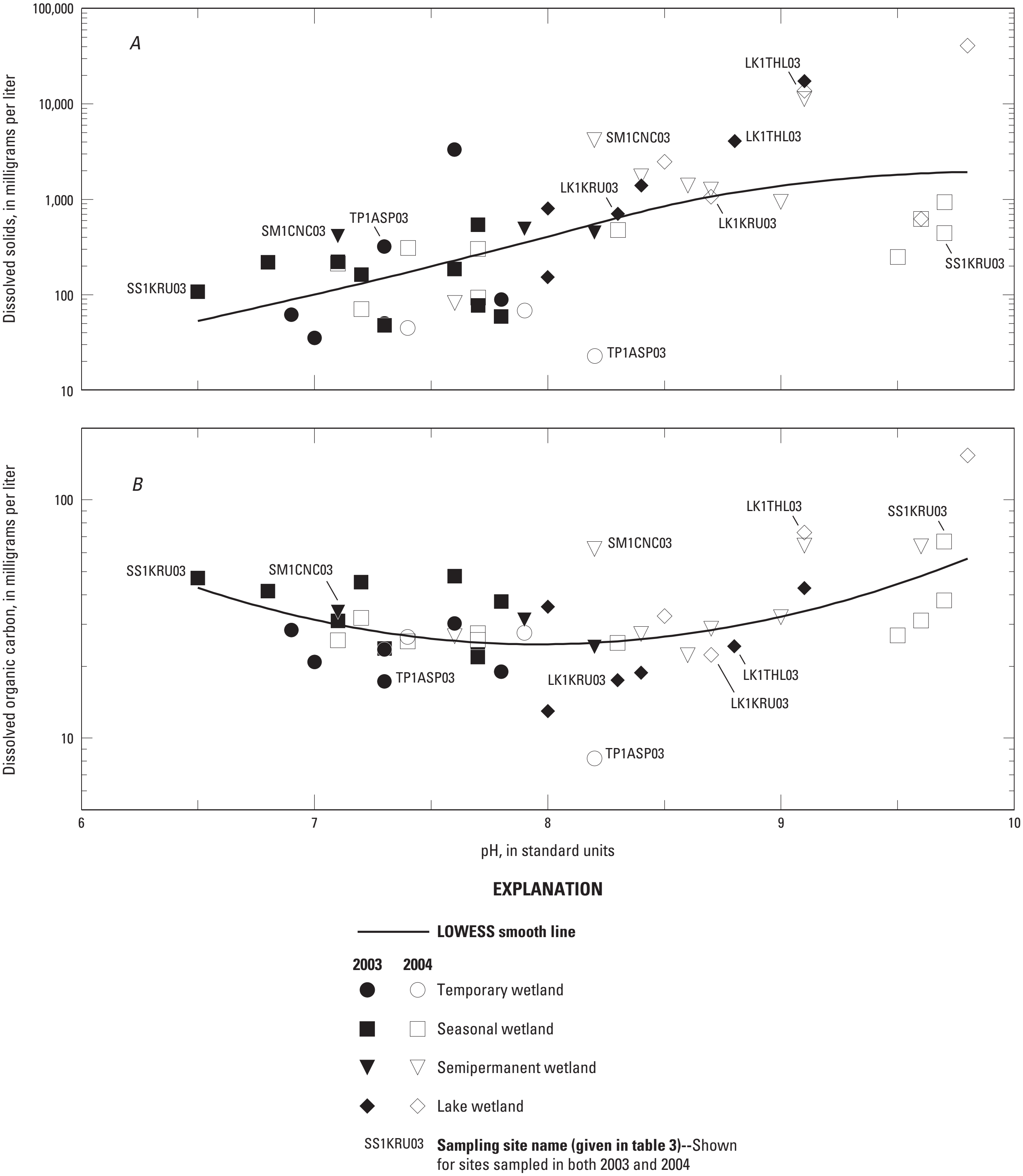


Figure 11. Relations between dissolved solids, dissolved organic carbon, selected mercury constituents, and pH in the Lostwood National Wildlife Refuge wetlands for 2003 and 2004.

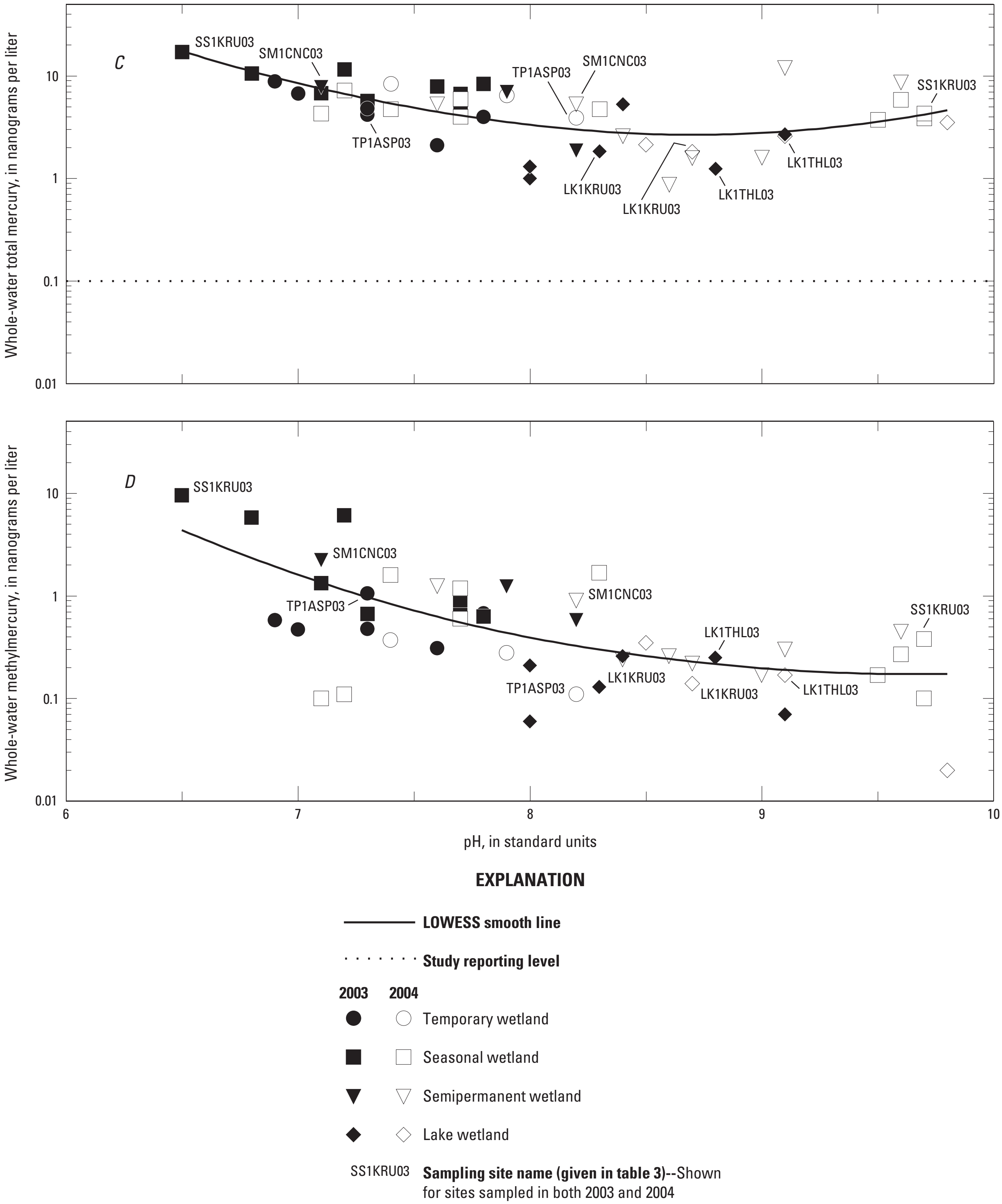


Figure 11. Relations between dissolved solids, dissolved organic carbon, selected mercury constituents, and pH in the Lostwood National Wildlife Refuge wetlands for 2003 and 2004.--Continued.

During winter, the depressional wetlands serve as accumulation points for snowfall. In the spring, the temporary, seasonal, and semipermanent wetlands are reflooded from snowmelt and spring rains (water of recent atmospheric origin and, thus, relatively low pH), and the soils rapidly evolve from aerobic to anaerobic conditions. Following reflooding, microbial process rates increase, using abundant labile organic carbon in sediments produced during the dessication period. Organic carbon use results in the release of DOC (consisting mostly of acidic humic substances; Thurman, 1985) to surface water. Runoff from adjacent terrestrial areas also can contribute DOC to the wetlands. The new DOC contributes to relatively low pH and enhances Hg solubility through complexation with organic ligands. With time, the humic substances are progressively metabolized and/or photochemically bleached to smaller and less complex aliphatic organic carbon compounds with low reactivity (Tulonen and others, 1992; Amon and Benner, 1996; del Giorgio and Cole, 1998; Miller, 1998; Waiser and Robarts, 2004). Associated with this process is an increase in pH as a result of several processes, including metabolic and photolytic alteration of the acidic humic substances, sulfate reduction (which generates bicarbonate and consumes hydronium ions), prolonged interaction with geologic materials, and common-ion effects of evaporative concentration (Banks and others, 2004). During the progressive metabolization, some of the DOC is incorporated into biomass and some is excreted or released back into the water. At some point, the availability of the degrading DOC for bacterial use is small, and evaporative concentration can increase DOC concentrations (Curtis and Adams, 1995; Waiser and Robarts, 2004). This probably contributed to the very large DOC concentrations (exceeding 50 mg/L) that occurred in some wetlands in 2004. Thus, although some exceptions exist, the general relation between DOC and pH is that relatively large DOC concentrations are associated with both relatively low and relatively high pH, and relatively small DOC concentrations are associated with intermediate pH. In the relatively low pH range, DOC probably is composed of compounds that have relatively high bioreactivity. In the relatively high pH range, DOC probably is composed of compounds that have relatively low bioreactivity. Patterns of DOC composition across the pH gradient probably affect patterns in both Hg and MeHg across the pH gradient.

Relatively large THg concentrations (greater than about 4 ng/L) generally were associated with both relatively low pH (values of less than about 8 standard units) and relatively high pH (values of more than about 9 standard units) (fig. 11*C*). Relatively large THg concentrations associated with relatively low pH typically occurred in temporary and seasonal wetlands although some also occurred in some semipermanent wetlands. Relatively small THg concentrations generally were associated with intermediate pH (values between about 8 and 9 standard units). The relation between THg (which generally consists mostly of inorganic Hg) and pH (fig. 11*C*) generally is similar to the relation between DOC and pH (fig. 11*B*) and indicates that solubility of inorganic Hg in the Refuge wetlands largely is affected by complexation with DOC. Within individual wetland types, relatively large THg concentrations also generally were associated with relatively large DOC concentrations (fig. 12), further supporting the role of DOC complexation in maintaining THg in the water column. Another factor that might affect the relation between THg and DOC is contributions of new water into the wetlands during snowmelt and runoff events. Because of the contributions of new DOC from the new water, the potential for the addition of new inorganic Hg is greater than during dry periods. Hg retained in the melting snowpack and on or in dead vegetation and oxidized soils might be mobilized during inundation and runoff. Thus, similar patterns in occurrence of THg and DOC across the pH gradient might also indicate that factors that affect differences in loading of new DOC among wetland types and between years also affect differences in loading of new inorganic Hg. During dry spring conditions, such as those that occurred in 2004, contributions of both new DOC and new inorganic Hg probably are small. However, some factors contribute to variability between patterns in THg and DOC across the pH gradient. DOC concentrations associated with high pH (that is, values of more than about 9.5 standard units) tend to be similar to or larger than those associated with low pH (values of less than about 7 standard units). Conversely, THg concentrations associated with high pH tend to be smaller than those associated with low pH. These patterns might indicate the occurrence of processes that result in removal of THg from the water column at a rate that exceeds effects of evaporative concentration on DOC. These processes might include semipermanent deposition in sediments and/or volatilization of Hg^0 to the atmosphere. Also, the specific composition of DOC at different pH ranges might affect the ability to complex with inorganic Hg. Thus, although some exceptions exist, the general relation between THg and pH is that relatively large THg concentrations are associated with both relatively low pH and relatively high pH. Relatively small THg concentrations generally are associated with intermediate pH. The relation between THg and pH generally is similar to the relation between DOC and pH and indicates an association between the occurrence of DOC and THg. Interactions between DOC and THg also affect the occurrence and production of MeHg.

Whole-water MeHg shows an inverse relation with pH (fig. 11*D*) and, thus, probably with residence time of water in the wetlands. In years that have substantial snowmelt or rainfall, the wetlands contain a higher proportion of water of recent atmospheric origin (and relatively low pH) with new DOC and new inorganic Hg than in other years. As the wetted soil conditions change from aerobic to anaerobic and when adequate sulfate is present, microbial sulfate reduction is stimulated and accompanying Hg methylation results. Thus, the drying and rewetting cycles appear to stimulate MeHg production in the Refuge wetlands. Characteristics of seasonal and semipermanent wetlands related to moderate frequency and duration of inundation appear to result in relatively large MeHg concentrations. Characteristics of temporary wetlands related to low frequency and short duration of inundation generally do not yield MeHg concentrations as large as those in seasonal and semipermanent wetlands. Lake wetlands also do not yield MeHg con-

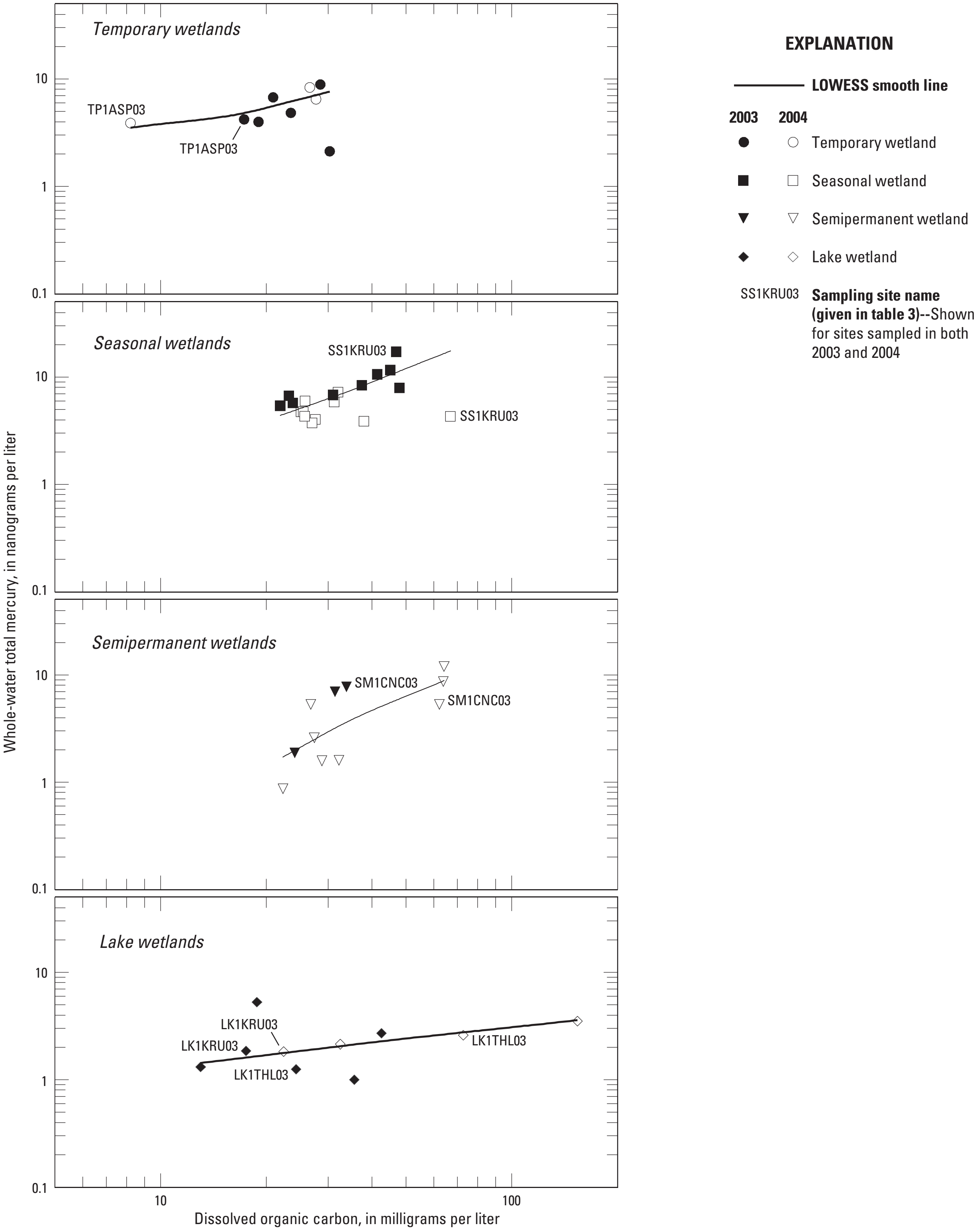


Figure 12. Relations between whole-water total mercury and dissolved organic carbon in the Lostwood National Wildlife Refuge wetlands for 2003 and 2004.

centrations as large as those in seasonal and semipermanent wetlands, partly because effects of drying cycles are less pronounced with less stimulation from oxidation of the sediments. Thus, although some exceptions exist, the general relation between MeHg and pH is that relatively large MeHg concentrations generally are associated with relatively low pH, and relatively small MeHg concentrations are associated with relatively high pH. Although the relation between MeHg and pH is relatively strong, pH probably does not have a direct dominant role in MeHg production. Rather, the interaction of several factors that occur across the pH gradient affect the resultant pattern.

Variability in dissolved-sulfate concentrations also can affect MeHg production. Relations between whole-water MeHg and dissolved sulfate for the various wetland types indicate the direct effects of sulfate concentrations on sulfate-reduction and Hg-methylation processes. Relations for seasonal and semipermanent wetlands (which generally had large MeHg concentrations) are shown in figure 13*A* for 2003 and in figure 13*B* for

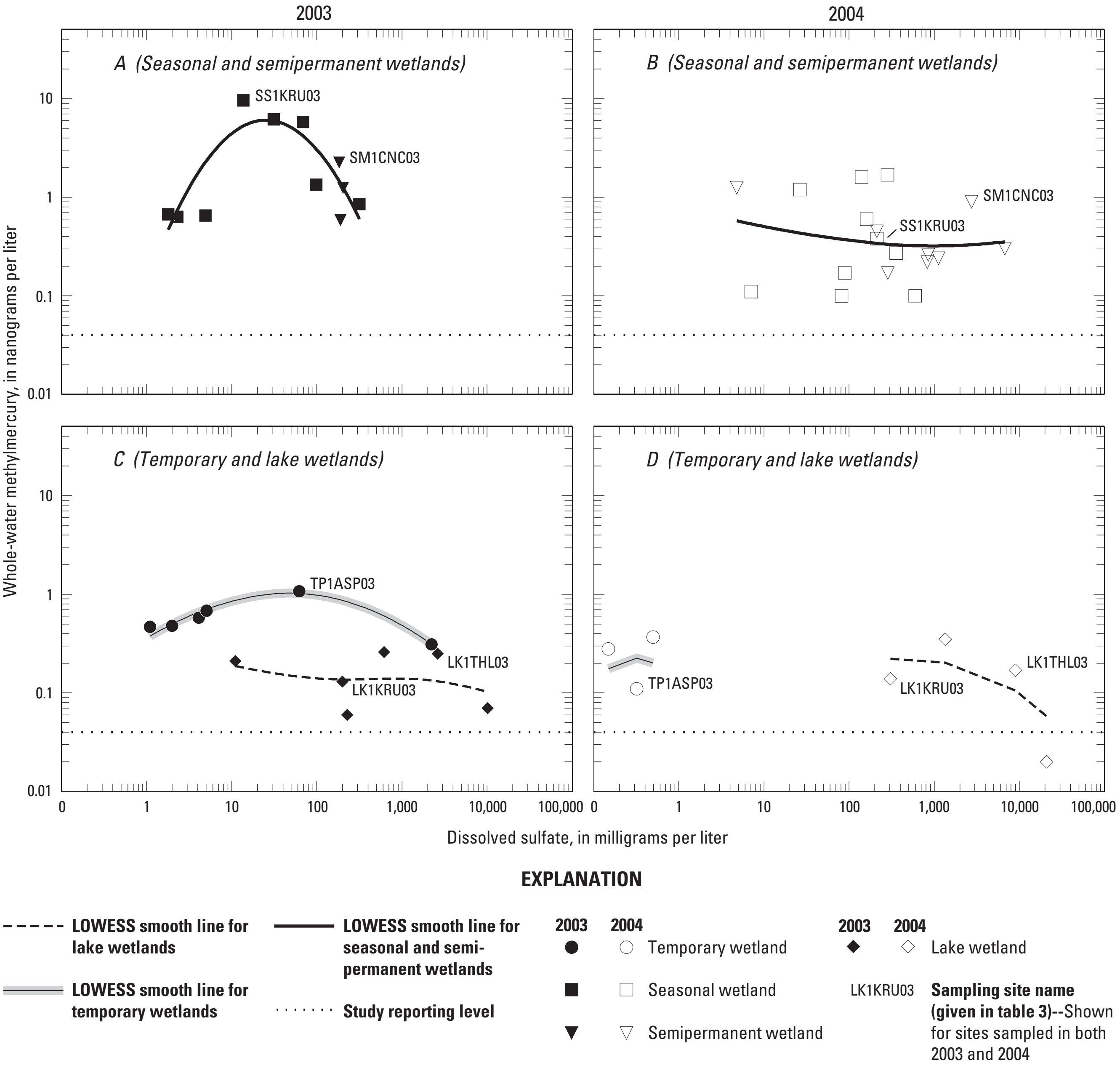


Figure 13. Relations between whole-water methylmercury and dissolved sulfate in the Lostwood National Wildlife Refuge wetlands for 2003 and 2004.

2004, and relations for temporary and lake wetlands (which generally had small MeHg concentrations) are shown in figure 13*C* for 2003 and in figure 13*D* for 2004. Relatively small sulfate concentrations result in relatively low rates of sulfate reduction; thus, little MeHg is produced (Gilmour and Krabbenhoft, 2001). Relatively large sulfate concentrations can result in high rates of sulfate reduction and accumulation of free sulfide in pore waters. Highly sulfidic environments appear to inhibit MeHg production although the mechanism by which that happens is not clear (Wiener and others, 2003). Intermediate sulfate concentrations have been shown to result in maximal MeHg production in other wetlands and stream ecosystems (Hurley and others, 1998; Krabbenhoft and others, 1999).

The largest MeHg concentrations (that is, greater than 1 ng/L) that occurred in the Refuge wetlands were associated with sulfate concentrations that ranged from about 5 to 280 mg/L. That range is a relatively narrow and intermediate interval at the low end of the overall range of sulfate concentrations in the Refuge wetlands (less than 0.30 to 21,000 mg/L). Concentrations in seasonal and semipermanent wetlands in 2003 (fig. 13*A*) show the convex relation between MeHg and dissolved sulfate commonly reported in other studies (U.S. Department of Commerce, National Oceanic and Atmospheric Administration, National Oceanic Service, 1996; Hurley and others, 1998; Krabbenhoft and others, 1999). However, the relation is not apparent in concentrations in seasonal and semipermanent wetlands in 2004. Several seasonal and semipermanent wetlands that had intermediate dissolved-sulfate concentrations (that is, between about 5 and 280 mg/L) also had relatively small MeHg concentrations (less than about 0.2 ng/L). Other factors, possibly relating to DOC processing or bioavailability of inorganic Hg, might have contributed to small MeHg concentrations in those wetlands. Most temporary wetlands had sulfate concentrations that were generally smaller than those in other wetland types (supplement 3) and also smaller than those in the range of concentrations associated with the largest MeHg concentrations (about 5 to 280 mg/L). Temporary wetlands also generally had smaller MeHg concentrations than seasonal and semipermanent wetlands (fig. 13). In some temporary wetlands, the relatively small sulfate concentrations might have limited MeHg production. Most lake wetlands had sulfate concentrations that were generally larger than those in other wetland types (supplement 3) and also larger than the range of concentrations associated with the largest MeHg concentrations. Lake wetlands also had generally smaller MeHg concentrations than other wetland types. Accumulation of free sulfide might have limited Hg methylation in some lake wetlands.

Examination of pH and concentrations of dissolved solids, dissolved sulfate, DOC, THg, and MeHg in 2003 and 2004 for seasonal wetland sampling site SS1KRU03 (specifically labeled in figures 11 and 12) provides evidence for possible explanations of associations among these variables in the Refuge wetlands. During the wet climatic conditions of 2003, SS1KRU03 had a relatively low pH of 6.5 standard units, a relatively small dissolved-solids concentration of 108 mg/L, an intermediate dissolved-sulfate concentration of 14 mg/L, a relatively large DOC concentration of 46.9 mg/L, a large THg concentration of 17.2 ng/L, and a large MeHg concentration of 9.6 ng/L. The relatively low pH, relatively small dissolved-solids concentration, and relatively large DOC concentration indicate the water in the wetland was predominantly of recent atmospheric origin and had abundant DOC with relatively high bioreactivity. The concomitant association of a large THg concentration probably indicates that processes that contributed both new DOC and new THg occurred and that substantial THg was maintained in the water column as a result of complexation with abundant DOC. The further concomitant association of a large MeHg concentration probably indicates that the abundant DOC with high bioreactivity was either being produced by or serving as a substrate for high rates of microbial activity (including sulfate reduction) and that the substantial THg was bioavailable to sulfate-reducing bacteria and was being actively methylated. The intermediate dissolved-sulfate concentration was adequate for sulfate reduction to occur but not large enough to promote excessive accumulation of free sulfide. During the dry climatic conditions of 2004, the pH value for SS1KRU03 had substantially increased to 9.7 standard units, the dissolved-solids concentration had increased to 442 mg/L, the dissolved-sulfate concentration had increased to 213 mg/L, the DOC concentration had moderately increased to 66.9 mg/L, the THg concentration had substantially decreased to 4.3 ng/L, and the MeHg concentration had very substantially decreased to 0.38 ng/L. The substantial increase in pH and dissolved solids between years indicates the wetland water was predominantly carry-over water as opposed to new water contributed from runoff or recent precipitation. The relatively large DOC concentration associated with the high pH and relatively large dissolved-solids concentrations indicates the DOC in the wetland water had been substantially bio- and photoprocessed (thus, microbial activity probably was substantially lower) and was affected by evaporative concentration. The fact that the increase in DOC was associated with a decrease in THg might indicate processes were occurring that resulted in removal of THg from the water column at a rate that exceeded effects of evaporative concentration on DOC. These processes might include semipermanent deposition in sediments and volatilization of Hg^0 to the atmosphere. However, the fact that the THg concentration in 2004 still was relatively large probably indicates the DOC concentration was able to maintain substantial THg in the water column. However, the concomitant association of a relatively small MeHg concentration (which represented about a 25-fold decrease from the 2003 MeHg concentration) indicates a much smaller proportion of THg (which probably was substantially affected by lower microbial activity) was being methylated in 2003 than in 2004. The relatively large dissolved-sulfate concentration in 2003 also might have contributed to accumulation of excessive free sulfide and reduced bioavailability of inorganic Hg; however, relatively large MeHg concentrations in some wetlands at the Refuge were associated with large dissolved-sulfate concentrations (as much as about 280 mg/L).

General water-quality characteristics of the Refuge wetlands vary substantially among wetland types and between years and result in notable patterns in occurrence of whole-water MeHg in the wetlands. Of the 44 wetlands sampled during this study, 11 had relatively large MeHg concentrations (that is, greater than 1 ng/L). Of these 11 wetlands, 7 were seasonal wetlands, 3 were semipermanent wetlands, and 1 was a temporary wetland. MeHg concentrations for three of the seasonal wetlands are considered very large (ranging from about 5 to 10 ng/L). Seasonal and semipermanent wetlands at the Refuge generally had larger MeHg concentrations than temporary and lake wetlands. All wetland types generally had substantially larger MeHg concentrations during the wet climatic conditions of 2003 than during the dry climatic conditions of 2004. Conditions associated with the largest whole-water MeHg concentrations (greater than 1 ng/L) include THg concentrations generally greater than about 4 ng/L, pH values less than about 8 standard units in conjunction with DOC concentrations generally greater than about 20 mg/L, and intermediate sulfate concentrations (generally between about 5 and 280 mg/L). These conditions generally were more prevalent in 2003 than in 2004 and were affected by the substantial snowmelt and runoff that occurred prior to sample collection. The distinct differences in MeHg concentrations between 2003 and 2004 indicate the importance of seemingly unrelated factors (such as climate) in regulating the formation of MeHg in the environment.

Summary and Conclusions

Certain ecosystem types, particularly wetlands, have environmental characteristics that make them particularly sensitive to mercury (Hg) inputs and that can result in large Hg concentrations in fish or other aquatic biota. However, wetlands vary considerably in their ecological characteristics, and many wetland types are understudied. Prairie pothole wetlands serve as a critical aquatic ecosystem for native and migratory wildlife in North America. However, little information is available on Hg cycling in the wetlands. Therefore, to provide the information needed to make effective management decisions to decrease human and wildlife exposure to methylmercury (MeHg), the U.S. Geological Survey, in cooperation with the North Dakota Department of Health, conducted a study to assess Hg and MeHg concentrations in prairie pothole wetlands at the Lostwood National Wildlife Refuge (the Refuge) in northwest North Dakota. In April 2003 and 2004, water and bottom-sediment samples were collected from 44 individual wetlands that were classified on the basis of water regime as one of four wetland types—temporary, seasonal, semipermanent, and lake. Many factors that may affect MeHg production, such as hydrology, surface-water chemistry, sediment chemistry, and Hg concentration, were considered in the study.

The prairie pothole wetlands at the Refuge had large ranges in major environmental characteristics. Hydrologic differences among wetland types, most notably semiannual wetting and drying cycles, that are intrinsic to prairie pothole wetlands, affected MeHg concentrations. This likely resulted from the stimulation of anaerobic microbial activity following reflooding of soils, particularly soils containing substantial organic carbon. Among the four wetland types considered for this study, seasonal and semipermanent wetlands generally had the largest MeHg concentrations. Lake wetlands, which do not undergo drying cycles, generally had the smallest MeHg concentrations. Temporary wetlands, which have the greatest frequency and longest duration of drying cycles, had intermediate MeHg concentrations that, in part, might reflect the limited amount of time (generally just a few weeks of inundation) these systems have to produce MeHg. Regardless of wetland type, however, MeHg concentrations at the Refuge are large in relation to reported concentrations for natural aquatic systems. Whole-water MeHg concentrations generally decrease with increases in pH, which generally is the relation reported in previously published Hg studies. The processing of organic carbon in soil and sediment is a major factor in regulating Hg cycling, especially MeHg production, in the environment. Dissolved organic carbon (DOC) concentrations at the Refuge are large (semipermanent and lake wetlands had DOC concentrations that exceeded 50 milligrams per liter), indicating the importance of organic carbon substrates for anaerobic processes occurring in those wetlands. DOC is mostly a byproduct of sediment organic carbon use and, because one important carbon-use process in those wetlands is sulfate reduction, the positive relation between MeHg and DOC was expected. The relation between whole-water THg (consisting mostly of inorganic Hg) and DOC indicates that solubility of inorganic Hg in the Refuge wetlands largely is controlled by complexation with DOC. Associated with the flux of water into the wetlands during spring are contributions of new DOC and the associated new inorganic Hg from the melting snowpack and rainfall. In both 2003 and 2004, MeHg had a convex relation with dissolved sulfate; the smallest MeHg concentrations were at the minimum and maximum sulfate concentrations. This complex relation is attributed to sulfate limitation effects at small sulfate concentrations and sulfide inhibition where sulfate concentrations are large and pore-water sulfide likely is elevated. Maximal MeHg concentrations in surface water were noted at intermediate sulfate concentrations (between about 5 and 280 milligrams per liter). Note that there are links between the intrinsic water chemistry and hydrology of the prairie pothole wetland types studied at the Refuge. Wetlands that had intermediate inundation periods (seasonal and semipermanent wetlands) have characteristic sulfate, DOC, and pH values that tend to promote maximal MeHg production response. However, all of the wetlands at the Refuge have large MeHg concentrations relative to most natural aquatic systems and, thus, should be considered as sensitive endpoints to Hg deposition.

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Supplements 1–6

Supplement 1: Quality Assurance/Quality Control

Quality-assurance/quality-control (QA/QC) samples collected during this study consisted of field-equipment blank and replicate samples to assess precision and accuracy of collected data. Blank samples provide information on potential effects of contamination during collection, processing, and laboratory analysis on analytical results. Replicate samples provide information on the precision or reproducibility of analytical results.

Quality-Assurance/Quality-Control Data Collection

Field-equipment blank samples were collected by passing ultra-pure water through the collection and processing equipment used for primary environmental samples. The procedures used to collect and process the samples were identical to those used for the environmental samples. A field-equipment blank sample with constituent concentrations equal to or less than the study reporting level for a given constituent indicates the overall process of sample collection, processing, and laboratory analysis is not introducing substantial contamination. Sporadic, infrequent detections at concentrations near the study reporting level probably represent random contamination or instrument calibration error that is not persistent in the process and that is not likely to cause substantial positive bias in study results. Also, consistent detections at concentrations that are substantially less than concentrations in environmental samples probably indicate routine contamination that does not substantially affect study results.

Replicate samples were used in this study to identify the level of precision (reproducibility) of analytical results. Replicate samples are two or more samples that are collected, processed, and analyzed in an identical manner. Replicate samples were collected and processed immediately after each associated primary environmental sample was collected and processed. Procedures used to collect and process the samples were identical to those used for the environmental samples.

Precision of analytical results for replicate samples is affected by numerous sources of variability potentially introduced by both field and laboratory processes, including sample collection, sample processing and handling, and laboratory preparation and analysis. Analyses of replicate samples, therefore, can indicate the reproducibility of environmental data and provide information on the adequacy of procedures to produce consistent results.

The precision of analytical results for the replicate samples was determined by calculating the standard deviation for each replicate pair as indicated in the following equation:

$$S = \sqrt{\frac{\sum d^2}{2}}, \qquad (1)$$

where

S is the standard deviation of the difference in concentration between replicate analyses, and

d is the difference in concentration between each pair of replicate analyses.

Precision also can be expressed as a relative standard deviation (RSD), in percent, which is computed from the standard deviation and the mean concentration for each replicate pair. Expressing precision relative to a mean concentration standardizes comparison of precision among individual constituents. The RSD, in percent, is calculated according to the following equation:

$$RSD = \frac{S}{X} * 100, \qquad (2)$$

where

RSD is the relative standard deviation,

S is the standard deviation, and

X is the mean concentration for a pair of replicate samples.

For equations 1 and 2, censored concentrations reported as less than the study reporting level were arbitrarily assigned a value of one-half the study reporting level.

Quality-Assurance/Quality-Control Results

Analytical results for field-equipment blank samples are given in supplement 5. Concentrations of most constituents in the field-equipment blank samples were less than or near the respective study analytical reporting level. Dissolved and suspended MeHg concentrations always were less than the study reporting levels. Dissolved solids, dissolved THg, suspended THg, whole-water THg, dissolved organic carbon, and whole-water organic carbon were either routinely or sporadically detected in the field-equipment blank samples but at concentrations less than the minimum environmental concentrations. The contamination for these constituents probably did not substantially affect study results. Dissolved sulfate and dissolved barium were routinely detected in field-equipment blank samples at small concentrations that were less than the concentrations of more than 90 percent of the environmental samples. The contamination for dissolved sulfate and dissolved barium probably did not substantially affect study results. Dissolved selenium was detected in a single field-equipment blank sample at a concentration within the range of typical environmental concentrations. However, because this contamination was sporadic and dissolved selenium was not determined to substantially relate to variability in any Hg constituents, the contamination probably did not substantially affect study results. In general, results for field-equipment blank samples indicate the overall process of sample collection, processing, and laboratory analysis did not introduce substantial contamination and did not substantially bias study results.

Analytical results for replicate samples are given in supplements 3 and 4, immediately following results for associated primary environmental samples. Statistical summaries of *RSD* values for pairs of primary environmental/replicate samples are given in supplement 6. Generally, an *RSD* value of 20 percent or less represents an acceptable level of precision although, for small concentrations near the limit of analytical detectability, the percent differences can be substantially larger and still be considered reasonable because of the limits of resolution (Taylor, 1987).

For this study, most constituents had *RSD* values less than 20 percent, indicating the overall process of sample collection, processing, and analysis was producing consistent results. Many of the *RSD* values were near 10 percent or less. However, several constituents, including carbonate, suspended solids, dissolved aluminum, dissolved chromium, dissolved copper, dissolved iron, dissolved manganese, dissolved selenium, dissolved zinc, dissolved and suspended MeHg, and suspended THg in water samples and MeHg and THg in bottom-sediment samples, had one or more replicate pairs with *RSD* values greater than 20 percent.

Carbonate, dissolved chromium, dissolved copper, dissolved selenium, and dissolved MeHg each had only one replicate pair with an *RSD* value that exceeded 20 percent. Those cases generally were for concentrations near the study reporting levels. Given the acceptable precision for most of the replicate pairs and consideration of limits of resolution at small concentrations, the precision results for these constituents are considered reasonable and acceptable. Dissolved manganese and dissolved zinc each had two replicate pairs with *RSD* values that exceeded 20 percent. Concentrations in the problematic replicate pairs for these constituents generally were near the study reporting level, and the precision results are considered reasonable and acceptable because of the limits of resolution at small concentrations. *RSD* values for suspended solids, dissolved aluminum, and dissolved iron showed consistently poor precision. Results for these constituents are given in supplements 3 and 4 but are not discussed elsewhere in this report because of the poor analytical precision and also because those constituents were not substantially relevant to the general findings of this study.

RSD values for suspended MeHg and suspended THg in water samples generally were greater than 20 percent. Concentrations for these constituents generally were small (generally less than 0.1 ng/L for suspended MeHg and 1 ng/L for suspended THg) and generally were near the study reporting levels where analytical resolution is poor. Given the relatively small amount of suspended material in the Refuge wetlands and the fact that suspended material generally is distributed more variably in natural waters than dissolved material, it is not unusual that the suspended THg and MeHg results would show relatively high *RSD* values. *RSD* values for whole-water MeHg and THg (calculated by summing the dissolved and suspended fractions of those constituents) generally were less than 20 percent. Thus, variability in the filtration process might have resulted in small differences in proportions of Hg being incorporated into either the dissolved or suspended samples. At the small concentrations that suspended Hg occurred in the samples, possible slight variability in the sample filtration process likely contributed to the relatively large *RSD* values for suspended Hg constituents. Given these factors, analytical results for suspended MeHg and suspended THg in water samples should be used with caution. However, the general findings of this study indicate the poor precision in the analytical results for suspended MeHg and suspended THg in water samples probably did not obscure patterns in differences in concentrations of those constituents among wetland types. In general, results for replicate samples indicate the overall process of sample collection, processing, and laboratory analysis provided accurate precision and did not substantially bias study results. Suspended MeHg and THg in bottom-sediment samples each had one replicate pair with *RSD* values that exceeded 20 percent. Given the acceptable precision for most of the replicate pairs and consideration of possible additional variability in the bottom-sediment sampling process, the precision results for those constituents are considered reasonable and acceptable.

Supplement 2. Water-quality property and constituent values.

[<less than; --, no data]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Depth to bottom from water surface at sampling site location (feet)	Barometric pressure (millimeters mercury)	Specific conductance (microsiemens per centimeter)	pH (standard units)	Air temperature (degrees Celsius)	Water temperature (degrees Celsius)	Turbidity (formazin nephelometric units)	Dissolved oxygen (milligrams per liter)
2003											
4	SS1THL03	4/21/2003	1620	1.1	700	151	7.7	22.0	16.9	28	12.3
6	SS1WLD03	4/22/2003	1530	1.0	705	843	7.7	20.5	14.0	<1.0	10.3
9	TP1WLD03	4/22/2003	1330	.5	705	97	7.3	20.5	15.2	<1.0	10.8
10	SM1WLD03	4/22/2003	1100	.9	705	736	8.2	17.0	10.8	3.2	8.1
11	TP1THL03	4/22/2003	0845	1.5	705	112	6.9	10.4	9.9	100	8.3
12	LK1THL03	4/22/2003	0930	3.9	703	11,600	8.8	6.9	7.4	18	16.6
18	SS3WDM03	4/24/2003	0830	1.1	700	359	6.8	14.6	10.3	60	5.1
20	SS2WDM03	4/24/2003	1110	1.4	700	290	7.2	16.4	11.7	35	6.1
22	SM1WDM03	4/24/2003	1405	1.1	700	744	7.9	24.0	14.7	30	8.6
22	SM1WDM03	4/24/2003	[1]1410	1.1	700	744	7.9	24.0	14.7	30	8.6
23	TP1CNC03	4/23/2003	1050	1.9	700	3,890	7.6	20.4	11.4	<1.0	10.5
25	SS4WDM03	4/24/2003	1630	2.1	698	378	7.1	24.0	15.6	20	8.7
26	SS1CNC03	4/24/2003	1120	.8	702	348	7.6	13.6	12.3	20	7.0
27	SM1CNC03	4/23/2003	0900	.8	695	310	7.1	16.8	8.6	61	4.3
28	LK1SEL03	4/23/2003	1100	3.3	695	298	8.0	18.6	11.9	40	9.3
28	LK1SEL03	4/23/2003	[1]1105	3.3	695	298	8.0	18.6	11.9	40	9.3
30	TP1ASP03	4/22/2003	1500	.7	702	511	7.3	20.5	17.0	4.0	5.5
31	TP1SEL03	4/23/2003	1430	1.7	695	67	7.0	23.8	16.4	14	9.8
33	LK1FSC03	4/23/2003	0740	4.3	696	1,280	8.0	15.0	11.1	40	7.0
34	LK1SSK03	4/24/2003	0950	2.6	702	2,090	8.4	13.2	12.6	110	8.2
36	LK1WWH03	4/24/2003	0810	3.3	702	22,500	9.1	12.2	10.8	5.7	6.4
38	SS1KRU03	4/24/2003	1440	1.0	698	217	6.5	20.4	10.2	18	1.3
39	SS2KRU03	4/24/2003	1350	1.3	700	87	7.3	14.8	14.8	5.0	8.9
40	LK1KRU03	4/22/2003	1330	5.6	704	1,150	8.3	19.8	12.3	6.0	11.8
41	TP1WSU03	4/23/2003	1250	.9	700	166	7.8	22.8	17.0	15	10.1
44	SS1ELN03	4/23/2003	1500	.7	695	109	7.8	21.7	18.1	40	11.1

Supplement 2. Water-quality property and constituent values.—Continued

[<less than; --, no data]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Depth to bottom from water surface at sampling site location (feet)	Barometric pressure (millimeters mercury)	Specific conductance (microsiemens per centimeter)	pH (standard units)	Air temperature (degrees Celsius)	Water temperature (degrees Celsius)	Turbidity (formazin nephelometric units)	Dissolved oxygen (milligrams per liter)
						2004					
1	SS4WLD04	4/28/2004	0800	1.6	679	355	7.1	9.2	11.5	3.7	7.3
1	SS4WLD04	4/28/2004	[1]0805	1.6	679	355	7.1	9.2	11.5	3.7	7.3
2	SM4WLD04	4/28/2004	0900	.7	680	1,080	8.7	8.4	10.1	<1.0	10.2
3	SM1THL04	4/28/2004	0800	1.3	682	1,730	8.6	8.9	11.4	6.9	8.3
5	SM3WLD04	4/28/2004	1130	.8	680	1,420	7.6	--	12.0	<1.0	9.1
7	TP2WLD04	4/28/2004	1115	.3	682	1,080	7.9	--	12.2	7.3	9.4
8	SM2WLD04	4/28/2004	1015	.3	682	720	9.6	--	12.0	1.3	11.5
12	LK1THL03	4/28/2004	1445	1.6	687	13,500	9.1	4.2	10.5	40	9.7
13	SS1TLS04	4/28/2004	1600	.7	690	743	8.3	2.3	9.1	61	10.0
14	SS1TWS04	4/26/2004	1245	2.3	707	735	9.6	8.6	15.0	27	14.4
15	SS2WLD04	4/27/2004	1255	.7	692	114	7.2	18.5	20.1	5.7	17.5
16	SS3WLD04	4/27/2004	1155	.7	693	371	7.4	16.0	14.2	4.7	10.8
16	SS3WLD04	4/27/2004	[1]1200	.7	693	371	7.4	16.0	14.2	4.7	10.8
17	SS1ASP04	4/27/2004	0830	.5	693	826	9.7	--	8.2	2.0	10.6
19	SM2CNC04	4/27/2004	1420	.7	692	2,050	8.4	18.5	18.6	3.9	11.0
21	TP2CNC04	4/26/2004	1555	1.0	704	48	7.4	10.2	14.2	7.4	12.3
24	SS3CNC04	4/27/2004	1500	1.3	690	143	7.7	--	18.5	2.5	10.1
27	SM1CNC03	4/28/2004	1430	1.3	688	4,590	8.2	--	9.8	50	12.9
29	LK1CNC04	4/28/2004	1220	3.3	683	3,210	8.5	7.0	12.4	11	8.6
30	TP1ASP03	4/26/2004	1435	1.0	705	40	8.2	11.1	13.7	4.8	9.7
32	SS1GNN04	4/27/2004	1530	.7	690	486	7.7	24.5	19.9	3.9	12.1
35	SM1KRU04	4/27/2004	1020	2.0	693	1,380	9.0	14.4	9.8	12	10.1
37	SS3KRU04	4/27/2004	0845	1.6	692	352	9.5	11.0	10.1	12	11.2
38	SS1KRU03	4/27/2004	0930	.7	692	627	9.7	13.8	10.0	55	13.6
40	LK1KRU03	4/28/2004	1050	2.6	--	10,600	8.7	9.4	11.8	34	14.2
42	SM1WLL04	4/28/2004	1520	.3	688	1,210	9.1	--	7.6	500	12.2
43	LK1ELS04	4/28/2004	0930	3.3	684	38,800	9.8	9.4	10.9	8.7	16.9

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[1] Quality-assurance/quality-control replicate sample.

Supplement 3. Major-ion, suspended-solids, and nonmercury trace-element data for water samples.

[mg/L, milligrams per liter; <, less than; °C, degrees Celsius; µg/L, micrograms per liter; U, analyzed for but not detected; M, no data]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Hardness, total, calculated (mg/L as $CaCO_3$)	Calcium, dissolved (mg/L)	Magnesium, dissolved (mg/L)	Sodium, dissolved (mg/L)	Percent sodium, in equivalents of major cations (percent)	Sodium adsorption ratio, calculated	Potassium, dissolved (mg/L)
					2003					
4	SS1THL03	4/21/2003	1620	49	11	5.1	<3.0	9	0.2	15
6	SS1WLD03	4/22/2003	1530	350	49	56	31	15	.7	23
9	TP1WLD03	4/22/2003	1330	28	7.1	2.4	<3.0	14	.2	10
10	SM1WLD03	4/22/2003	1100	310	43	50	24	13	.6	20
11	TP1THL03	4/22/2003	0845	26	4.8	3.3	<3.0	12	.3	18
12	LK1THL03	4/22/2003	0930	2,000	12	470	560	37	6.0	100
18	SS3WDM03	4/24/2003	0830	140	30	16	4.7	6	.2	21
20	SS2WDM03	4/24/2003	1110	98	21	11	3.1	5	.1	31
22	SM1WDM03	4/24/2003	1405	340	75	37	16	8	.4	35
22	SM1WDM03	4/24/2003	[1]1410	340	74	37	16	8	.4	35
23	TP1CNC03	4/23/2003	1050	2,200	360	330	170	14	2.0	5.7
25	SS4WDM03	4/24/2003	1630	110	25	10	<3.0	4	.1	46
26	SS1CNC03	4/24/2003	1120	120	31	10	<3.0	4	.1	39
27	SM1CNC03	4/23/2003	0900	200	39	25	54	34	2.0	21
28	LK1SEL03	4/23/2003	1100	140	28	16	4.6	6	.2	11
28	LK1SEL03	4/23/2003	[1]1105	130	27	16	4.8	7	.2	11
30	TP1ASP03	4/22/2003	1500	290	78	24	4.9	3	.1	6.6
31	TP1SEL03	4/23/2003	1430	17	4.0	1.6	<3.0	19	.3	8.5
33	LK1FSC03	4/23/2003	0740	480	53	84	96	28	2.0	43
34	LK1SSK03	4/24/2003	0950	370	25	75	360	66	8.0	24
36	LK1WWH03	4/24/2003	0810	860	6.7	200	5,500	92	81	190
38	SS1KRU03	4/24/2003	1440	46	8.8	5.8	<3.0	7	.2	32
39	SS2KRU03	4/24/2003	1350	20	4.4	2.2	<3.0	15	.3	13
40	LK1KRU03	4/22/2003	1330	300	21	60	150	50	4.0	17
41	TP1WSU03	4/23/2003	1250	56	14	4.9	7.4	19	.4	10
44	SS1ELN03	4/23/2003	1500	20	4.2	2.3	<3.0	12	.3	20

Supplement 3. Major-ion, suspended-solids, and nonmercury trace-element data for water samples.—Continued

[mg/L, milligrams per liter; <, less than; °C, degrees Celsius; µg/L, micrograms per liter; U, analyzed for but not detected; M, no data]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Hardness, total, calculated (mg/L as $CaCO_3$)	Calcium, dissolved (mg/L)	Magnesium, dissolved (mg/L)	Sodium, dissolved (mg/L)	Percent sodium, in equivalents of major cations (percent)	Sodium adsorption ratio, calculated	Potassium, dissolved (mg/L)
2004										
1	SS4WLD04	4/28/2004	0800	150	29	20	7.2	8	0.2	13
1	SS4WLD04	4/28/2004	[1]0805	150	28	20	7.3	8	.3	13
2	SM4WLD04	4/28/2004	0900	800	130	110	44	10	.7	54
3	SM1THL04	4/28/2004	0800	870	100	150	93	18	1.0	29
5	SM3WLD04	4/28/2004	1130	56	13	5.6	3.2	9	.2	12
7	TP2WLD04	4/28/2004	1115	47	11	4.6	<3.0	10	.2	8.3
8	SM2WLD04	4/28/2004	1015	350	74	40	20	9	.5	67
12	LK1THL03	4/28/2004	1445	6,500	16	1,600	2,000	39	11	350
13	SS1TLS04	4/28/2004	1600	290	53	39	22	13	.6	21
14	SS1TWS04	4/26/2004	1245	440	120	34	10	4	.2	19
15	SS2WLD04	4/27/2004	1255	39	8.6	4.3	<3.0	10	.2	15
16	SS3WLD04	4/27/2004	1155	220	53	22	4.2	4	.1	16
16	SS3WLD04	4/27/2004	[1]1200	220	54	22	4.4	4	.1	17
17	SS1ASP04	4/27/2004	0830	600	150	52	22	7	.4	45
19	SM2CNC04	4/27/2004	1420	1,000	140	170	110	18	2.0	48
21	TP2CNC04	4/26/2004	1555	16	3.1	1.9	<3.0	18	.3	11
24	SS3CNC04	4/27/2004	1500	60	14	5.9	<3.0	8	.2	9.9
27	SM1CNC03	4/28/2004	1430	2,100	420	250	410	29	4.0	100
29	LK1CNC04	4/28/2004	1220	930	40	200	470	51	7.0	43
30	TP1ASP03	4/26/2004	1435	13	3.7	1.0	<3.0	29	.4	1.9
32	SS1GNN04	4/27/2004	1530	200	40	24	9.7	8	.3	20
35	SM1KRU04	4/27/2004	1020	290	29	53	230	61	6.0	20
37	SS3KRU04	4/27/2004	0845	180	44	17	4.1	4	.1	15
38	SS1KRU03	4/27/2004	0930	270	57	30	17	10	.5	45
40	LK1KRU03	4/28/2004	1050	460	40	86	220	49	4.0	26
42	SM1WLL04	4/28/2004	1520	2,000	60	450	3,000	74	29	190
43	LK1ELS04	4/28/2004	0930	400	5.7	94	15,000	98	320	300

Supplement 3. Major-ion, suspended-solids, and nonmercury trace-element data for water samples.—Continued

[mg/L, milligrams per liter; <, less than; °C, degrees Celsius; µg/L, micrograms per liter; U, analyzed for but not detected; M, no data]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Acid neutralizing capacity, whole-water, titration to pH 4.5, laboratory (mg/L as $CaCO_3$)	Bicarbonate, whole-water, titration to pH 4.5, laboratory (mg/L)	Carbonate, whole-water, titration to pH 8.3, laboratory (mg/L)	Hydroxide, whole-water, titration to pH 10.4, laboratory (mg/L)	Sulfate, dissolved (mg/L)	Chloride, dissolved (mg/L)
2003									
4	SS1THL03	4/21/2003	1620	56	68	<1	<1	4.9	2.1
6	SS1WLD03	4/22/2003	1530	100	122	<1	<1	320	9.3
9	TP1WLD03	4/22/2003	1330	38	46	<1	<1	2.0	1.0
10	SM1WLD03	4/22/2003	1100	185	226	<1	<1	190	5.9
11	TP1THL03	4/22/2003	0845	39	47	<1	<1	4.1	3.4
12	LK1THL03	4/22/2003	0930	393	286	95	<1	2,600	72
18	SS3WDM03	4/24/2003	0830	119	145	<1	<1	69	5.4
20	SS2WDM03	4/24/2003	1110	96	118	<1	<1	31	5.7
22	SM1WDM03	4/24/2003	1405	182	222	<1	<1	200	9.2
22	SM1WDM03	4/24/2003	[1]1410	175	214	<1	<1	200	9.0
23	TP1CNC03	4/23/2003	1050	407	497	<1	<1	2,200	13
25	SS4WDM03	4/24/2003	1630	47	57	<1	<1	98	8.8
26	SS1CNC03	4/24/2003	1120	124	151	<1	<1	24	2.2
27	SM1CNC03	4/23/2003	0900	136	166	<1	<1	180	7.7
28	LK1SEL03	4/23/2003	1100	133	161	1	<1	11	1.8
28	LK1SEL03	4/23/2003	[1]1105	139	169	<1	<1	12	1.8
30	TP1ASP03	4/22/2003	1500	239	291	<1	<1	62	1.9
31	TP1SEL03	4/23/2003	1430	24	29	<1	<1	1.1	.9
33	LK1FSC03	4/23/2003	0740	474	562	8	<1	230	15
34	LK1SSK03	4/24/2003	0950	503	542	35	<1	620	16
36	LK1WWH03	4/24/2003	0810	1,870	2,280	<1	<1	10,200	220
38	SS1KRU03	4/24/2003	1440	63	77	<1	<1	14	4.0
39	SS2KRU03	4/24/2003	1350	32	39	<1	<1	1.8	1.7
40	LK1KRU03	4/22/2003	1330	414	451	27	<1	200	10
41	TP1WSU03	4/23/2003	1250	72	88	<1	<1	5.1	2.9
44	SS1ELN03	4/23/2003	1500	38	46	<1	<1	2.3	2.6

Supplement 3. Major-ion, suspended-solids, and nonmercury trace-element data for water samples.—Continued

[mg/L, milligrams per liter; <, less than; °C, degrees Celsius; µg/L, micrograms per liter; U, analyzed for but not detected; M, no data]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Acid neutralizing capacity, whole-water, titration to pH 4.5, laboratory (mg/L as $CaCO_3$)	Bicarbonate, whole-water, titration to pH 4.5, laboratory (mg/L)	Carbonate, whole-water, titration to pH 8.3, laboratory (mg/L)	Hydroxide, whole-water, titration to pH 10.4, laboratory (mg/L)	Sulfate, dissolved (mg/L)	Chloride, dissolved (mg/L)
2004									
1	SS4WLD04	4/28/2004	0800	96	117	<1	<1	82	1.7
1	SS4WLD04	4/28/2004	[1]0805	96	117	<1	<1	84	1.7
2	SM4WLD04	4/28/2004	0900	112	89	23	<1	830	15
3	SM1THL04	4/28/2004	0800	208	222	16	<1	850	14
5	SM3WLD04	4/28/2004	1130	66	80	<1	<1	4.8	.9
7	TP2WLD04	4/28/2004	1115	63	77	<1	<1	<.30	.6
8	SM2WLD04	4/28/2004	1015	298	187	87	<1	210	15
12	LK1THL03	4/28/2004	1445	1,230	939	280	<1	9,100	17
13	SS1TLS04	4/28/2004	1600	86	105	<1	<1	280	6.8
14	SS1TWS04	4/26/2004	1245	131	90	34	<1	360	3.6
15	SS2WLD04	4/27/2004	1255	48	59	<1	<1	7.1	1.4
16	SS3WLD04	4/27/2004	1155	112	137	<1	<1	140	3.0
16	SS3WLD04	4/27/2004	[1]1200	113	138	<1	<1	140	2.7
17	SS1ASP04	4/27/2004	0830	83	30	35	<1	600	9.8
19	SM2CNC04	4/27/2004	1420	191	224	4	<1	1,100	24
21	TP2CNC04	4/26/2004	1555	37	45	<1	<1	.50	.7
24	SS3CNC04	4/27/2004	1500	50	60	<1	<1	26	1.4
27	SM1CNC03	4/28/2004	1430	280	320	11	<1	2,800	31
29	LK1CNC04	4/28/2004	1220	583	673	19	<1	1,400	25
30	TP1ASP03	4/26/2004	1435	18	22	<1	<1	.30	<.3
32	SS1GNN04	4/27/2004	1530	71	86	<1	<1	160	2.2
35	SM1KRU04	4/27/2004	1020	508	521	48	<1	290	9.3
37	SS3KRU04	4/27/2004	0845	132	96	32	<1	90	1.1
38	SS1KRU03	4/27/2004	0930	121	67	40	<1	210	5.0
40	LK1KRU03	4/28/2004	1050	612	637	54	<1	310	16
42	SM1WLL04	4/28/2004	1520	876	752	156	<1	6,800	150
43	LK1ELS04	4/28/2004	0930	6,580	2,560	2,690	<1	21,000	370

Supplement 3. Major-ion, suspended-solids, and nonmercury trace-element data for water samples.—Continued

[mg/L, milligrams per liter; <, less than; °C, degrees Celsius; µg/L, micrograms per liter; U, analyzed for but not detected; M, no data]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Solids, dissolved, sum of constituents (mg/L)	Solids, suspended, residue on evaporation at 105°C (mg/L)	Aluminum, dissolved (µg/L)	Antimony, dissolved (µg/L)	Arsenic, dissolved (µg/L)	Barium, dissolved (µg/L)
2003									
4	SS1THL03	4/21/2003	1620	77	6	<50	<1	<1	49.8
6	SS1WLD03	4/22/2003	1530	545	<5	<50	<1	<1	41.4
9	TP1WLD03	4/22/2003	1330	50	11	80	<1	<1	25.1
10	SM1WLD03	4/22/2003	1100	445	<5	60	<1	2	55.6
11	TP1THL03	4/22/2003	0845	62	8	200	<1	<1	23.3
12	LK1THL03	4/22/2003	0930	4,090	9	<50	<1	14	6.8
18	SS3WDM03	4/24/2003	0830	218	6	<50	<1	1	74.9
20	SS2WDM03	4/24/2003	1110	163	<5	200	<1	1	71.1
22	SM1WDM03	4/24/2003	1405	488	48	500	<1	2	66.7
22	SM1WDM03	4/24/2003	[1]1410	480	29	400	<1	2	64.9
23	TP1CNC03	4/23/2003	1050	3,340	6	<50	<1	2	55.6
25	SS4WDM03	4/24/2003	1630	222	118	200	<1	2	51.0
26	SS1CNC03	4/24/2003	1120	185	<5	<50	<1	3	74.7
27	SM1CNC03	4/23/2003	0900	412	9	<50	<1	<1	68.8
28	LK1SEL03	4/23/2003	1100	153	8	200	<1	1	45.9
28	LK1SEL03	4/23/2003	[1]1105	157	<5	<50	<1	1	39.1
30	TP1ASP03	4/22/2003	1500	323	9	<50	<1	1	117
31	TP1SEL03	4/23/2003	1430	35	<5	<50	<1	<1	2.6
33	LK1FSC03	4/23/2003	0740	803	<5	<50	<1	4	70.8
34	LK1SSK03	4/24/2003	0950	1,410	21	200	<1	6	38.3
36	LK1WWH03	4/24/2003	0810	17,400	6	<50	<1	42	<1.0
38	SS1KRU03	4/24/2003	1440	108	10	<50	<1	<1	53.0
39	SS2KRU03	4/24/2003	1350	48	<5	<50	<1	<1	3.0
40	LK1KRU03	4/22/2003	1330	709	12	80	<1	3	25.8
41	TP1WSU03	4/23/2003	1250	90	5	<50	<1	<1	21.0
44	SS1ELN03	4/23/2003	1500	59	<5	<50	<1	<1	3.8

Supplement 3. Major-ion, suspended-solids, and nonmercury trace-element data for water samples.—Continued

[mg/L, milligrams per liter; <, less than; °C, degrees Celsius; µg/L, micrograms per liter; U, analyzed for but not detected; M, no data]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Solids, dissolved, sum of constituents (mg/L)	Solids, suspended, residue on evaporation at 105°C (mg/L)	Aluminum, dissolved (µg/L)	Antimony, dissolved (µg/L)	Arsenic, dissolved (µg/L)	Barium, dissolved (µg/L)
2004									
1	SS4WLD04	4/28/2004	0800	212	<5	100	<1	1	70.0
1	SS4WLD04	4/28/2004	[1]0805	214	<5	70	<1	1	74.3
2	SM4WLD04	4/28/2004	0900	1,260	<5	80	<1	3	105
3	SM1THL04	4/28/2004	0800	1,370	10	<50	<1	4	54.0
5	SM3WLD04	4/28/2004	1130	81	<5	<50	<1	1	53.7
7	TP2WLD04	4/28/2004	1115	68	<5	50	<1	<1	32.4
8	SM2WLD04	4/28/2004	1015	610	412	6,800	<1	12	226
12	LK1THL03	4/28/2004	1445	13,800	23	80	2	67	23.6
13	SS1TLS04	4/28/2004	1600	479	8	80	<1	2	81.3
14	SS1TWS04	4/26/2004	1245	625	33	1,800	<1	13	62.4
15	SS2WLD04	4/27/2004	1255	71	<5	60	<1	1	32.3
16	SS3WLD04	4/27/2004	1155	309	<5	60	<1	1	59.1
16	SS3WLD04	4/27/2004	[1]1200	306	8	100	<1	1	50.0
17	SS1ASP04	4/27/2004	0830	935	15	<50	<1	2	82.5
19	SM2CNC04	4/27/2004	1420	1,740	<5	<50	<1	5	82.8
21	TP2CNC04	4/26/2004	1555	45	8	70	<1	1	13.4
24	SS3CNC04	4/27/2004	1500	93	<5	<50	<1	2	47.0
27	SM1CNC03	4/28/2004	1430	4,130	27	400	<1	14	105
29	LK1CNC04	4/28/2004	1220	2,480	10	<50	<1	16	25.1
30	TP1ASP03	4/26/2004	1435	23	11	80	<1	<1	23.0
32	SS1GNN04	4/27/2004	1530	303	<5	<50	<1	<1	70.1
35	SM1KRU04	4/27/2004	1020	934	23	200	<1	4	46.7
37	SS3KRU04	4/27/2004	0845	251	16	1,300	<1	4	93.0
38	SS1KRU03	4/27/2004	0930	442	19	200	<1	7	44.7
40	LK1KRU03	4/28/2004	1050	1,060	50	500	<1	7	67.0
42	SM1WLL04	4/28/2004	1520	11,100	194	2,500	4	36	89.1
43	LK1ELS04	4/28/2004	0930	40,700	16	<50	<1	520	26.1

Supplement 3. Major-ion, suspended-solids, and nonmercury trace-element data for water samples.—Continued

[mg/L, milligrams per liter; <, less than; °C, degrees Celsius; µg/L, micrograms per liter; U, analyzed for but not detected; M, no data]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Beryllium, dissolved (µg/L)	Boron, dissolved (µg/L)	Cadmium, dissolved (µg/L)	Chromium, dissolved (µg/L)	Copper, dissolved (µg/L)	Iron, dissolved (µg/L)	Lead, dissolved (µg/L)
2003										
4	SS1THL03	4/21/2003	1620	<1	60	<1	<1	<1	60	<1
6	SS1WLD03	4/22/2003	1530	<1	90	<1	<1	<1	80	<1
9	TP1WLD03	4/22/2003	1330	<1	<50	<1	<1	<1	140	<1
10	SM1WLD03	4/22/2003	1100	<1	80	<1	<1	<1	140	<1
11	TP1THL03	4/22/2003	0845	<1	50	<1	<1	<1	140	<1
12	LK1THL03	4/22/2003	0930	<1	380	<1	<1	<1	150	<1
18	SS3WDM03	4/24/2003	0830	<1	80	<1	<1	2	200	<1
20	SS2WDM03	4/24/2003	1110	<1	100	<1	<1	<1	410	<1
22	SM1WDM03	4/24/2003	1405	<1	60	<1	<1	<1	850	<1
22	SM1WDM03	4/24/2003	[1]1410	<1	60	<1	<1	<1	660	<1
23	TP1CNC03	4/23/2003	1050	<1	310	<1	3	1	320	<1
25	SS4WDM03	4/24/2003	1630	<1	100	<1	<1	1	550	<1
26	SS1CNC03	4/24/2003	1120	<1	160	<1	<1	2	130	<1
27	SM1CNC03	4/23/2003	0900	<1	110	<1	<1	<1	90	<1
28	LK1SEL03	4/23/2003	1100	<1	90	<1	<1	<1	720	<1
28	LK1SEL03	4/23/2003	[1]1105	<1	70	<1	3	<1	260	<1
30	TP1ASP03	4/22/2003	1500	<1	60	<1	<1	<1	200	<1
31	TP1SEL03	4/23/2003	1430	<1	60	<1	<1	<1	50	<1
33	LK1FSC03	4/23/2003	0740	<1	<50	<1	<1	<1	50	<1
34	LK1SSK03	4/24/2003	0950	<1	310	<1	<1	2	320	<1
36	LK1WWH03	4/24/2003	0810	<1	400	<1	<1	15	100	<1
38	SS1KRU03	4/24/2003	1440	<1	90	<1	<1	2	260	<1
39	SS2KRU03	4/24/2003	1350	<1	70	<1	<1	<1	30	<1
40	LK1KRU03	4/22/2003	1330	<1	170	<1	<1	<1	210	<1
41	TP1WSU03	4/23/2003	1250	<1	U	<1	<1	2	40	<1
44	SS1ELN03	4/23/2003	1500	<1	60	<1	<1	<1	60	<1

Supplement 3. Major-ion, suspended-solids, and nonmercury trace-element data for water samples.—Continued

[mg/L, milligrams per liter; <, less than; °C, degrees Celsius; µg/L, micrograms per liter; U, analyzed for but not detected; M, no data]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Beryllium, dissolved (µg/L)	Boron, dissolved (µg/L)	Cadmium, dissolved (µg/L)	Chromium, dissolved (µg/L)	Copper, dissolved (µg/L)	Iron, dissolved (µg/L)	Lead, dissolved (µg/L)
2004										
1	SS4WLD04	4/28/2004	0800	<1	50	<1	<1	<1	210	<1
1	SS4WLD04	4/28/2004	[1]0805	<1	60	<1	<1	<1	160	<1
2	SM4WLD04	4/28/2004	0900	<1	60	<1	<1	2	120	<1
3	SM1THL04	4/28/2004	0800	<1	70	<1	<1	M	80	<1
5	SM3WLD04	4/28/2004	1130	<1	50	<1	<1	<1	240	<1
7	TP2WLD04	4/28/2004	1115	<1	60	<1	<1	1	100	<1
8	SM2WLD04	4/28/2004	1015	<1	70	<1	9	10	7,200	7
12	LK1THL03	4/28/2004	1445	<1	1,300	<1	1	50	308	<1
13	SS1TLS04	4/28/2004	1600	<1	90	<1	<1	1	120	<1
14	SS1TWS04	4/26/2004	1245	<1	190	<1	3	3	1,600	2
15	SS2WLD04	4/27/2004	1255	<1	70	<1	<1	<1	140	<1
16	SS3WLD04	4/27/2004	1155	<1	60	<1	<1	<1	130	<1
16	SS3WLD04	4/27/2004	[1]1200	<1	60	<1	<1	1	210	<1
17	SS1ASP04	4/27/2004	0830	<1	<50	<1	<1	2	40	<1
19	SM2CNC04	4/27/2004	1420	<1	90	<1	<1	3	70	<1
21	TP2CNC04	4/26/2004	1555	<1	50	<1	<1	1	60	<1
24	SS3CNC04	4/27/2004	1500	<1	50	<1	<1	<1	90	<1
27	SM1CNC03	4/28/2004	1430	<1	260	<1	1	10	600	<1
29	LK1CNC04	4/28/2004	1220	<1	290	<1	<1	10	160	<1
30	TP1ASP03	4/26/2004	1435	<1	U	<1	<1	1	70	<1
32	SS1GNN04	4/27/2004	1530	<1	70	<1	<1	1	100	<1
35	SM1KRU04	4/27/2004	1020	<1	190	<1	<1	5	380	<1
37	SS3KRU04	4/27/2004	0845	<1	110	<1	2	3	1,000	2
38	SS1KRU03	4/27/2004	0930	<1	190	<1	<1	2	500	<1
40	LK1KRU03	4/28/2004	1050	<1	190	<1	1	6	1,300	1
42	SM1WLL04	4/28/2004	1520	<1	1,100	<1	5	70	4,200	2
43	LK1ELS04	4/28/2004	0930	<1	2,500	<1	50	590	200	<1

Supplement 3. Major-ion, suspended-solids, and nonmercury trace-element data for water samples.—Continued

[mg/L, milligrams per liter; <, less than; °C, degrees Celsius; µg/L, micrograms per liter; U, analyzed for but not detected; M, no data]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Manganese, dissolved (µg/L)	Nickel, dissolved (µg/L)	Selenium, dissolved (µg/L)	Silver, dissolved (µg/L)	Thallium, dissolved (µg/L)	Zinc, dissolved (µg/L)
2003									
4	SS1THL03	4/21/2003	1620	<10	1	<1	<1	<1.0	<1
6	SS1WLD03	4/22/2003	1530	70	2	<1	<1	<1.0	<1
9	TP1WLD03	4/22/2003	1330	<10	<1	<1	<1	<1.0	2
10	SM1WLD03	4/22/2003	1100	110	2	<1	<1	<1.0	<1
11	TP1THL03	4/22/2003	0845	<10	<1	<1	<1	<1.0	2
12	LK1THL03	4/22/2003	0930	<10	<1	<1	<1	<1.0	<1
18	SS3WDM03	4/24/2003	0830	180	1	<1	<1	<1.0	1
20	SS2WDM03	4/24/2003	1110	40	1	<1	<1	<1.0	3
22	SM1WDM03	4/24/2003	1405	210	3	<1	<1	<1.0	3
22	SM1WDM03	4/24/2003	[1]1410	200	3	<1	<1	<1.0	2
23	TP1CNC03	4/23/2003	1050	<10	11	2	<1	<1.0	3
25	SS4WDM03	4/24/2003	1630	40	2	<1	<1	<1.0	4
26	SS1CNC03	4/24/2003	1120	<10	2	<1	<1	<1.0	9
27	SM1CNC03	4/23/2003	0900	20	2	<1	<1	<1.0	1
28	LK1SEL03	4/23/2003	1100	50	1	<1	<1	<1.0	<1
28	LK1SEL03	4/23/2003	[1]1105	20	1	<1	<1	<1.0	<1
30	TP1ASP03	4/22/2003	1500	20	2	2	<1	<1.0	<1
31	TP1SEL03	4/23/2003	1430	<10	<1	<1	<1	<1.0	<1
33	LK1FSC03	4/23/2003	0740	<10	3	<1	<1	<1.0	<1
34	LK1SSK03	4/24/2003	0950	<10	2	1	<1	<1.0	2
36	LK1WWH03	4/24/2003	0810	<10	20	<1	<1	<1.0	<1
38	SS1KRU03	4/24/2003	1440	180	<1	<1	<1	<1.0	20
39	SS2KRU03	4/24/2003	1350	<10	<1	<1	<1	<1.0	4
40	LK1KRU03	4/22/2003	1330	<10	1	<1	<1	<1.0	<1
41	TP1WSU03	4/23/2003	1250	10	<1	<1	<1	<1.0	<1
44	SS1ELN03	4/23/2003	1500	<10	<1	<1	<1	<1.0	3

Supplement 3. Major-ion, suspended-solids, and nonmercury trace-element data for water samples.—Continued

[mg/L, milligrams per liter; <, less than; °C, degrees Celsius; µg/L, micrograms per liter; U, analyzed for but not detected; M, no data]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Manganese, dissolved (µg/L)	Nickel, dissolved (µg/L)	Selenium, dissolved (µg/L)	Silver, dissolved (µg/L)	Thallium, dissolved (µg/L)	Zinc, dissolved (µg/L)
2004									
1	SS4WLD04	4/28/2004	0800	70	1	1	<1	<1.0	2
1	SS4WLD04	4/28/2004	[1]0805	60	2	<1	<1	<1.0	3
2	SM4WLD04	4/28/2004	0900	60	5	3	<1	<1.0	4
3	SM1THL04	4/28/2004	0800	70	M	4	<1	<1.0	3
5	SM3WLD04	4/28/2004	1130	40	1	<1	<1	<1.0	2
7	TP2WLD04	4/28/2004	1115	30	1	<1	<1	<1.0	8
8	SM2WLD04	4/28/2004	1015	340	12	4	<1	<1.0	50
12	LK1THL03	4/28/2004	1445	30	3	23	<1	<1.0	20
13	SS1TLS04	4/28/2004	1600	40	2	5	<1	<1.0	3
14	SS1TWS04	4/26/2004	1245	160	7	13	<1	<1.0	20
15	SS2WLD04	4/27/2004	1255	30	1	1	<1	<1.0	7
16	SS3WLD04	4/27/2004	1155	20	2	<1	<1	<1.0	2
16	SS3WLD04	4/27/2004	[1]1200	30	2	<1	<1	<1.0	2
17	SS1ASP04	4/27/2004	0830	20	6	5	<1	<1.0	2
19	SM2CNC04	4/27/2004	1420	40	5	10	<1	<1.0	5
21	TP2CNC04	4/26/2004	1555	20	<1	3	<1	<1.0	4
24	SS3CNC04	4/27/2004	1500	10	<1	6	<1	<1.0	3
27	SM1CNC03	4/28/2004	1430	620	16	11	<1	<1.0	8
29	LK1CNC04	4/28/2004	1220	50	2	8	<1	<1.0	4
30	TP1ASP03	4/26/2004	1435	10	<1	1	<1	<1.0	2
32	SS1GNN04	4/27/2004	1530	<10	2	<1	<1	<1.0	2
35	SM1KRU04	4/27/2004	1020	70	2	3	<1	<1.0	3
37	SS3KRU04	4/27/2004	0845	170	4	<1	<1	<1.0	30
38	SS1KRU03	4/27/2004	0930	210	5	<1	<1	<1.0	4
40	LK1KRU03	4/28/2004	1050	130	3	5	<1	<1.0	5
42	SM1WLL04	4/28/2004	1520	200	14	33	<1	<1.0	20
43	LK1ELS04	4/28/2004	0930	30	<1	28	<1	<1.0	70

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[1] Quality-assurance/quality-control replicate sample.

Supplement 4. Mercury and organic-carbon data for water and bottom-sediment samples.

[ng/L, nanograms per liter; E, estimated; --, no data; <, less than; mg/L, milligrams per liter; ng/g, nanograms per gram]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Methylmercury, dissolved (ng/L)	Methylmercury, suspended (ng/L)	Methylmercury, whole-water (ng/L)	Inorganic mercury, dissolved (ng/L)	Inorganic mercury, suspended (ng/L)	Inorganic mercury, whole-water (ng/L)
2003									
4	SS1THL03	4/21/2003	1620	0.58	0.07	0.65	5.58	0.45	6.03
6	SS1WLD03	4/22/2003	1530	.75	.10	.85	3.97	.58	4.55
9	TP1WLD03	4/22/2003	1330	.43	.05	.48	4.12	.23	4.35
10	SM1WLD03	4/22/2003	1100	.52	.06	.58	.86	.44	1.30
11	TP1THL03	4/22/2003	0845	.54	.04	.58	7.90	.40	8.30
12	LK1THL03	4/22/2003	0930	.14	.11	.25	.56	.44	1.00
18	SS3WDM03	4/24/2003	0830	5.76	.07	5.80	3.71	.65	4.80
20	SS2WDM03	4/24/2003	1110	E5.91	.22	E6.13	E5.29	.16	E5.47
22	SM1WDM03	4/24/2003	1405	.86	.38	1.24	1.52	E4.23	E5.75
22	SM1WDM03	4/24/2003	[1]1410	1.07	.14	1.21	.85	1.54	2.39
23	TP1CNC03	4/23/2003	1050	.25	.06	.31	1.42	.39	1.81
25	SS4WDM03	4/24/2003	1630	1.28	.05	1.33	5.14	.33	5.47
26	SS1CNC03	4/24/2003	1120	.44	--	--	6.62	--	--
27	SM1CNC03	4/23/2003	0900	E2.16	.08	E2.24	E5.23	.27	E5.50
28	LK1SEL03	4/23/2003	1100	.18	.03	.21	.54	.56	1.10
28	LK1SEL03	4/23/2003	[1]1105	.12	--	--	.68	--	--
30	TP1ASP03	4/22/2003	1500	.90	.18	1.07	2.16	.96	3.13
31	TP1SEL03	4/23/2003	1430	.40	.04	.47	5.60	1.06	6.25
33	LK1FSC03	4/23/2003	0740	.05	<.03	E.05	.72	E.23	E.95
34	LK1SSK03	4/24/2003	0950	.21	.05	.26	.80	4.22	5.02
36	LK1WWH03	4/24/2003	0810	.04	.03	.07	1.70	.95	2.64
38	SS1KRU03	4/24/2003	1440	E9.16	.40	E9.56	E5.44	2.17	E7.64
39	SS2KRU03	4/24/2003	1350	.53	.14	.67	4.79	.27	5.06
40	LK1KRU03	4/22/2003	1330	.10	.03	.13	.68	1.04	1.72
41	TP1WSU03	4/23/2003	1250	.61	.07	.68	3.01	.29	3.30
44	SS1ELN03	4/23/2003	1500	.59	.04	.63	7.12	.64	7.76

Supplement 4. Mercury and organic-carbon data for water and bottom-sediment samples.—Continued

[ng/L, nanograms per liter; E, estimated; --, no data; <, less than; mg/L, milligrams per liter; ng/g, nanograms per gram]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Methylmercury, dissolved (ng/L)	Methylmercury, suspended (ng/L)	Methylmercury, whole-water (ng/L)	Inorganic mercury, dissolved (ng/L)	Inorganic mercury, suspended (ng/L)	Inorganic mercury, whole-water (ng/L)
2004									
1	SS4WLD04	4/28/2004	0800	0.10	<0.03	E0.10	4.20	<0.10	E4.20
1	SS4WLD04	4/28/2004	[1]0805	.09	.04	.13	4.38	<.10	E4.34
2	SM4WLD04	4/28/2004	0900	.19	.03	.22	1.39	<.10	E1.36
3	SM1THL04	4/28/2004	0800	.26	<.03	E.26	.61	<.10	E.61
5	SM3WLD04	4/28/2004	1130	1.22	.03	1.25	3.94	.11	4.05
7	TP2WLD04	4/28/2004	1115	.28	<.03	E.28	5.96	.23	E6.20
8	SM2WLD04	4/28/2004	1015	.15	.30	.45	2.67	5.55	8.22
12	LK1THL03	4/28/2004	1445	.07	.10	.17	.53	1.89	2.42
13	SS1TLS04	4/28/2004	1600	1.62	.06	1.68	2.77	E.29	E3.06
14	SS1TWS04	4/26/2004	1245	<.04	.27	E.27	E3.84	1.74	E5.58
15	SS2WLD04	4/27/2004	1255	<.04	.11	E.11	E6.69	E.43	E7.12
16	SS3WLD04	4/27/2004	1155	1.57	.03	1.60	2.81	E.34	E3.15
16	SS3WLD04	4/27/2004	[1]1200	1.19	.14	1.33	3.29	E.12	E3.41
17	SS1ASP04	4/27/2004	0830	.06	.04	.10	2.64	E1.13	E3.77
19	SM2CNC04	4/27/2004	1420	.17	.07	.24	1.54	E.82	E2.36
21	TP2CNC04	4/26/2004	1555	.29	.08	.37	7.31	E.67	E7.98
24	SS3CNC04	4/27/2004	1500	1.19	<.03	E1.19	4.79	<.10	E4.79
27	SM1CNC03	4/28/2004	1430	.69	.21	.90	3.30	E1.10	E4.40
29	LK1CNC04	4/28/2004	1220	.29	.06	.35	1.22	E.56	E1.79
30	TP1ASP03	4/26/2004	1435	.08	.03	.11	3.13	E.65	E3.78
32	SS1GNN04	4/27/2004	1530	.50	.10	.60	3.53	<.10	E3.43
35	SM1KRU04	4/27/2004	1020	.11	.06	.17	1.25	E.18	E1.43
37	SS3KRU04	4/27/2004	0845	.10	.07	.17	2.82	E.75	E3.57
38	SS1KRU03	4/27/2004	0930	.22	.16	.38	3.50	E.41	E3.91
40	LK1KRU03	4/28/2004	1050	.07	.07	.14	.73	E.96	E1.69
42	SM1WLL04	4/28/2004	1520	.06	.24	.30	2.58	9.13	11.7
43	LK1ELS04	4/28/2004	0930	<.04	.03	<.04	E2.80	E.69	E3.50

Supplement 4. Mercury and organic-carbon data for water and bottom-sediment samples.—Continued

[ng/L, nanograms per liter; E, estimated; --, no data; <, less than; mg/L, milligrams per liter; ng/g, nanograms per gram]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Total mercury, dissolved (ng/L)	Total mercury, suspended (ng/L)	Total mercury, whole-water (ng/L)	Ratio of whole-water methylmercury to whole-water total mercury	Organic carbon, dissolved (mg/L)	Organic carbon, whole-water (mg/L)
2003									
4	SS1THL03	4/21/2003	1620	6.16	0.52	6.68	0.097	23.2	23.2
6	SS1WLD03	4/22/2003	1530	4.72	.68	5.40	.157	21.9	22.5
9	TP1WLD03	4/22/2003	1330	4.55	.28	4.83	.099	23.5	23.6
10	SM1WLD03	4/22/2003	1100	1.38	.50	1.88	.309	24.1	24.4
11	TP1THL03	4/22/2003	0845	8.44	.44	8.88	.065	28.5	29.0
12	LK1THL03	4/22/2003	0930	.70	.55	1.25	.200	24.3	30.6
18	SS3WDM03	4/24/2003	0830	9.47	.72	10.6	.547	41.4	42.4
20	SS2WDM03	4/24/2003	1110	11.20	.38	11.6	E.528	45.1	41.6
22	SM1WDM03	4/24/2003	1405	2.38	E4.61	E6.99	E.180	31.4	32.1
22	SM1WDM03	4/24/2003	[1]1410	1.92	1.68	3.60	.336	31.1	31.6
23	TP1CNC03	4/23/2003	1050	1.67	.45	2.12	.146	30.3	30.7
25	SS4WDM03	4/24/2003	1630	6.42	.38	6.80	.196	31.0	32.3
26	SS1CNC03	4/24/2003	1120	7.06	.86	7.92	--	47.9	48.6
27	SM1CNC03	4/23/2003	0900	7.39	.35	7.74	E.289	33.8	33.5
28	LK1SEL03	4/23/2003	1100	.72	.59	1.31	.160	13.0	13.3
28	LK1SEL03	4/23/2003	[1]1105	.80	.32	1.12	--	13.1	13.7
30	TP1ASP03	4/22/2003	1500	3.06	1.14	4.20	.255	17.3	17.9
31	TP1SEL03	4/23/2003	1430	6.00	1.10	6.72	.070	20.9	21.2
33	LK1FSC03	4/23/2003	0740	.77	.23	1.00	E.060	35.6	36.4
34	LK1SSK03	4/24/2003	0950	1.01	4.27	5.28	.049	18.8	18.4
36	LK1WWH03	4/24/2003	0810	1.74	.98	2.71	.026	42.6	40.4
38	SS1KRU03	4/24/2003	1440	14.60	2.57	17.2	E.556	46.9	41.4
39	SS2KRU03	4/24/2003	1350	5.32	.41	5.73	.117	23.8	22.9
40	LK1KRU03	4/22/2003	1330	.78	1.07	1.85	.070	17.5	17.0
41	TP1WSU03	4/23/2003	1250	3.62	.36	3.98	.171	19.0	19.7
44	SS1ELN03	4/23/2003	1500	7.71	.68	8.39	.075	37.4	38.6

Supplement 4. Mercury and organic-carbon data for water and bottom-sediment samples.—Continued

[ng/L, nanograms per liter; E, estimated; --, no data; <, less than; mg/L, milligrams per liter; ng/g, nanograms per gram]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Total mercury, dissolved (ng/L)	Total mercury, suspended (ng/L)	Total mercury, whole-water (ng/L)	Ratio of whole-water methylmercury to whole-water total mercury	Organic carbon, dissolved (mg/L)	Organic carbon, whole-water (mg/L)
2004									
1	SS4WLD04	4/28/2004	0800	4.30	<0.10	E4.30	E0.023	25.7	24.4
1	SS4WLD04	4/28/2004	[1]0805	4.47	<.10	E4.47	E.029	25.3	24.0
2	SM4WLD04	4/28/2004	0900	1.58	<.10	E1.58	E.139	28.8	28.8
3	SM1THL04	4/28/2004	0800	.87	<.10	E.87	E.299	22.3	21.9
5	SM3WLD04	4/28/2004	1130	5.16	E.14	E5.30	E.236	26.8	26.2
7	TP2WLD04	4/28/2004	1115	6.24	E.24	E6.48	E.043	27.7	28.4
8	SM2WLD04	4/28/2004	1015	2.82	5.85	8.67	.052	63.8	64.8
12	LK1THL03	4/28/2004	1445	.60	1.99	2.59	.066	72.8	72.7
13	SS1TLS04	4/28/2004	1600	4.39	E.35	E4.74	E.354	25.1	25.1
14	SS1TWS04	4/26/2004	1245	3.84	2.01	5.85	E.046	31.2	30.7
15	SS2WLD04	4/27/2004	1255	6.69	E.54	E7.23	E.015	32.0	32.0
16	SS3WLD04	4/27/2004	1155	4.38	E.37	E4.75	E.337	25.5	25.1
16	SS3WLD04	4/27/2004	[1]1200	4.48	E.26	E4.74	E.281	25.2	24.7
17	SS1ASP04	4/27/2004	0830	2.70	E1.17	E3.87	E.026	37.9	38.4
19	SM2CNC04	4/27/2004	1420	1.71	E.89	E2.60	E.092	27.4	27.5
21	TP2CNC04	4/26/2004	1555	7.60	E.75	E8.35	E.044	26.6	25.5
24	SS3CNC04	4/27/2004	1500	5.98	<.10	E5.98	E.199	25.8	25.1
27	SM1CNC03	4/28/2004	1430	3.99	E1.31	E5.30	E.170	62.2	62.6
29	LK1CNC04	4/28/2004	1220	1.51	E.62	E2.14	E.166	32.5	31.3
30	TP1ASP03	4/26/2004	1435	3.21	E.68	E3.89	E.028	8.2	8.2
32	SS1GNN04	4/27/2004	1530	4.03	<.10	E4.03	E.149	27.6	27.7
35	SM1KRU04	4/27/2004	1020	1.36	E.24	E1.60	E.106	32.2	31.2
37	SS3KRU04	4/27/2004	0845	2.92	E.82	E3.74	E.045	27.0	28.2
38	SS1KRU03	4/27/2004	0930	3.72	E.57	E4.29	E.089	66.9	68.1
40	LK1KRU03	4/28/2004	1050	.80	E1.03	E1.83	E.077	22.4	23.6
42	SM1WLL04	4/28/2004	1520	2.64	9.37	12.0	.025	64.2	66.2
43	LK1ELS04	4/28/2004	0930	2.80	E.72	E3.52	E.006	154	164

Supplement 4. Mercury and organic-carbon data for water and bottom-sediment samples.—Continued

[ng/L, nanograms per liter; E, estimated; --, no data; <, less than; mg/L, milligrams per liter; ng/g, nanograms per gram]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Methylmercury, sediment (ng/g)	Total mercury, sediment (ng/g)	Ratio of methylmercury to total mercury, sediment	Sediment, dry weight (percent of wet weight)	Loss on ignition, sediment, dry weight (percent)
				2003				
4	SS1THL03	4/21/2003	1620	0.51	53.9	0.009	33.4	29.5
6	SS1WLD03	4/22/2003	1530	.10	30.2	.007	39.8	23.5
9	TP1WLD03	4/22/2003	1330	.61	40.7	.015	44.2	16.9
10	SM1WLD03	4/22/2003	1100	.08	51.2	.004	25.4	32.0
11	TP1THL03	4/22/2003	0845	.14	34.8	.006	43.2	18.2
12	LK1THL03	4/22/2003	0930	.05	18.2	.011	37.5	11.0
18	SS3WDM03	4/24/2003	0830	.58	60.3	.010	43.1	19.5
20	SS2WDM03	4/24/2003	1110	.75	48.4	.015	45.4	17.6
22	SM1WDM03	4/24/2003	1405	1.17	60.8	.019	21.4	35.4
22	SM1WDM03	4/24/2003	[1]1410	1.02	68.1	.015	20.6	36.8
23	TP1CNC03	4/23/2003	1050	.26	28.0	.007	42.8	19.0
25	SS4WDM03	4/24/2003	1630	2.05	50.5	.041	27.7	24.4
26	SS1CNC03	4/24/2003	1120	3.28	55.3	.059	25.3	34.9
27	SM1CNC03	4/23/2003	0900	.25	40.8	.005	44.1	19.3
28	LK1SEL03	4/23/2003	1100	.60	59.7	.010	20.1	28.9
28	LK1SEL03	4/23/2003	[1]1105	.71	47.1	.015	21.0	30.2
30	TP1ASP03	4/22/2003	1500	1.28	73.0	.018	16.4	45.0
31	TP1SEL03	4/23/2003	1430	.66	25.7	.026	54.5	11.6
33	LK1FSC03	4/23/2003	0740	.53	49.9	.011	28.1	23.5
34	LK1SSK03	4/24/2003	0950	.07	20.1	.010	45.8	8.08
36	LK1WWH03	4/24/2003	0810	.03	6.77	.030	68.0	2.84
38	SS1KRU03	4/24/2003	1440	1.12	38.4	.029	36.5	18.2
39	SS2KRU03	4/24/2003	1350	.84	58.5	.014	27.4	31.2
40	LK1KRU03	4/22/2003	1330	.08	23.3	.009	37.7	10.3
41	TP1WSU03	4/23/2003	1250	.16	27.5	.007	69.1	5.72
44	SS1ELN03	4/23/2003	1500	.58	35.9	.016	44.6	16.6

Supplement 4. Mercury and organic-carbon data for water and bottom-sediment samples.—Continued

[ng/L, nanograms per liter; E, estimated; --, no data; <, less than; mg/L, milligrams per liter; ng/g, nanograms per gram]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Methylmercury, sediment (ng/g)	Total mercury, sediment (ng/g)	Ratio of methylmercury to total mercury, sediment	Sediment, dry weight (percent of wet weight)	Loss on ignition, sediment, dry weight (percent)
2004								
1	SS4WLD04	4/28/2004	0800	2.57	48.6	0.053	26.4	33.7
1	SS4WLD04	4/28/2004	[1]0805	4.72	64.7	.073	21.0	44.8
2	SM4WLD04	4/28/2004	0900	<.39	59.8	.003	20.3	34.0
3	SM1THL04	4/28/2004	0800	<.31	42.4	.005	25.6	30.3
5	SM3WLD04	4/28/2004	1130	.34	38.8	.005	40.8	18.8
7	TP2WLD04	4/28/2004	1115	1.30	99.0	.013	26.2	29.6
8	SM2WLD04	4/28/2004	1015	.53	43.2	.012	20.6	33.4
12	LK1THL03	4/28/2004	1445	<.11	8.54	.023	73.5	1.30
13	SS1TLS04	4/28/2004	1600	1.05	38.3	.027	29.6	30.9
14	SS1TWS04	4/26/2004	1245	<.22	22.7	.009	35.6	8.90
15	SS2WLD04	4/27/2004	1255	.43	22.0	.020	41.4	18.6
16	SS3WLD04	4/27/2004	1155	2.07	34.4	.060	30.5	23.9
16	SS3WLD04	4/27/2004	[1]1200	1.64	18.8	.087	36.1	18.4
17	SS1ASP04	4/27/2004	0830	1.13	58.0	.019	15.9	47.5
19	SM2CNC04	4/27/2004	1420	2.54	58.9	.043	15.0	51.0
21	TP2CNC04	4/26/2004	1555	.46	51.2	.009	39.4	23.3
24	SS3CNC04	4/27/2004	1500	4.16	89.4	.047	24.0	42.4
27	SM1CNC03	4/28/2004	1430	.95	54.4	.017	25.4	31.7
29	LK1CNC04	4/28/2004	1220	<.14	11.2	.017	58.1	5.80
30	TP1ASP03	4/26/2004	1435	1.82	71.3	.026	27.0	39.1
32	SS1GNN04	4/27/2004	1530	1.77	68.6	.026	23.2	36.5
35	SM1KRU04	4/27/2004	1020	.60	41.6	.014	16.6	42.0
37	SS3KRU04	4/27/2004	0845	1.58	41.6	.038	29.7	32.6
38	SS1KRU03	4/27/2004	0930	3.36	40.0	.084	29.7	24.5
40	LK1KRU03	4/28/2004	1050	.50	26.9	.019	32.2	20.3
42	SM1WLL04	4/28/2004	1520	<.19	30.7	.007	41.3	15.5
43	LK1ELS04	4/28/2004	0930	<.14	8.66	.023	56.1	6.00

[1] Quality-assurance/quality-control replicate sample.

Supplement 5. Analytical results for quality-assurance/quality-control field-equipment blank samples.

[ng/L, nanograms per liter; mg/L, milligrams per liter; <, less than; µS/cm, microsiemens per centimeter; ANC, acid neutralizing capacity; °C, degrees Celsius; µg/L, micrograms per liter; U, analyzed for but not detected]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Methylmercury, dissolved (ng/L)	Methylmercury, suspended (ng/L)	Total mercury, dissolved (ng/L)	Total mercury, suspended (ng/L)	Organic carbon, dissolved (mg/L)	Organic carbon, whole-water (mg/L)
Mercury and organic carbon (U.S. Geological Survey Wisconsin Water Science Center Mercury Laboratory, Madison, Wisconsin)									
2003									
18	SS3WDM03	4/24/2003	0825	<0.04	<0.027	0.50	0.147	0.05	0.09
33	LK1FSC03	4/23/2003	0735	<.04	<.027	.09	<.068	.01	.01
2004									
14	SS1TWS04	4/26/2004	1250	<0.04	<0.027	0.48	<0.068	0.22	0.23
17	SS1ASP04	4/27/2004	0835	<.04	<.027	.34	<.068	.46	.18

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Specific conductance, laboratory (µS/cm)	Calcium, dissolved (mg/L)	Magnesium, dissolved (mg/L)	Potassium, dissolved (mg/L)	Sodium, dissolved (mg/L)	ANC, whole-water, titration to pH 4.5, laboratory (mg/L as $CaCO_3$)	Bicarbonate, whole-water, titration to pH 4.5, laboratory (mg/L)	Carbonate, whole-water, titration to pH 8.3, laboratory (mg/L)
Inorganic properties and constituents (North Dakota Department of Health Laboratory, Bismarck, North Dakota)											
2003											
18	SS3WDM03	4/24/2003	0825	1	<2.00	<1.00	<1.00	<3.00	2	2	<1
33	LK1FSC03	4/23/2003	0735	1	<2.00	<1.00	<1.00	<3.00	2	2	<1
2004											
14	SS1TWS04	4/26/2004	1250	3	<2.00	<1.00	<1.00	<3.00	5	6	<1
17	SS1ASP04	4/27/2004	0835	2	<2.00	<1.00	<1.00	<3.00	4	5	<1

Supplement 5. Analytical results for quality-assurance/quality-control field-equipment blank samples.—Continued

[ng/L, nanograms per liter; mg/L, milligrams per liter; <, less than; µS/cm, microsiemens per centimeter; ANC, acid neutralizing capacity; °C, degrees Celsius; µg/L, micrograms per liter; U, analyzed for but not detected]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Hydroxide, whole-water, titration to pH 10.4, laboratory (mg/L)	Sulfate, dissolved (mg/L)	Chloride, dissolved (mg/L)	Solids, dissolved, sum of constituents (mg/L)	Solids, suspended, residue on evaporation at 105°C (mg/L)	Aluminum, dissolved (µg/L)	Antimony, dissolved (µg/L)	Arsenic, dissolved (µg/L)
Inorganic properties and constituents, Continued (North Dakota Department of Health Laboratory, Bismarck, North Dakota)											
2003											
18	SS3WDM03	4/24/2003	0825	<1	1.3	<0.3	12	<5.00	<50	<1	<1.0
33	LK1FSC03	4/23/2003	0735	<1	1	<.3	11	<5.00	<50	<1	<1.0
2004											
14	SS1TWS04	4/26/2004	1250	<1	0.4	<0.3	13	<5.00	<50	<1	<1.0
17	SS1ASP04	4/27/2004	0835	<1	<.3	<.3	12	<5.00	<50	<1	1.3

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Barium, dissolved (µg/L)	Beryllium, dissolved (µg/L)	Boron, dissolved (µg/L))	Cadmium, dissolved (µg/L)	Chromium, dissolved (µg/L)	Copper, dissolved (µg/L)	Iron, dissolved (µg/L)	Lead, dissolved (µg/L)
Inorganic properties and constituents, Continued (North Dakota Department of Health Laboratory, Bismarck, North Dakota)											
2003											
18	SS3WDM03	4/24/2003	0825	<1.0	<1	U	<1	<1	<1	<10	<1
33	LK1FSC03	4/23/2003	0735	<1.0	<1	<50	<1	<1	<1	<10	<1
2004											
14	SS1TWS04	4/26/2004	1250	8.1	<1	<50	<1	<1	<1	10	<1
17	SS1ASP04	4/27/2004	0835	4.2	<1	<50	<1	<1	<1	10	<1

Supplement 5. Analytical results for quality-assurance/quality-control field-equipment blank samples.—Continued

[ng/L, nanograms per liter; mg/L, milligrams per liter; <, less than; µS/cm, microsiemens per centimeter; ANC, acid neutralizing capacity; °C, degrees Celsius; µg/L, micrograms per liter; U, analyzed for but not detected]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Manganese, dissolved (µg/L)	Nickel, dissolved (µg/L)	Selenium, dissolved (µg/L)	Silver, dissolved (µg/L)	Thallium, dissolved (µg/L)	Zinc, dissolved (µg/L)
Inorganic properties and constituents, Continued (North Dakota Department of Health Laboratory, Bismarck, North Dakota)									
2003									
18	SS3WDM03	4/24/2003	0825	<10	<1	<1	<1	<1.0	<1
33	LK1FSC03	4/23/2003	0735	<10	<1	<1	<1	<1.0	<1
2004									
14	SS1TWS04	4/26/2004	1250	<10	<1	<1	<1	<1.0	1
17	SS1ASP04	4/27/2004	0835	<10	<1	4	<1	<1.0	1

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Specific conductance, laboratory (µS/cm)	Calcium, dissolved (mg/L)	Magnesium, dissolved (mg/L)	Potassium, dissolved (mg/L)	Sodium, dissolved (mg/L)	ANC, whole-water, titration to pH 4.5, laboratory (mg/L as $CaCO_3$)	Bicarbonate, whole-water, titration to pH 4.5, laboratory (mg/L)
Inorganic properties, major ions, and suspended solids (North Dakota Department of Health Laboratory, Bismarck, North Dakota)										
2003										
18	SS3WDM03	4/24/2003	0825	1	<2.00	<1.00	<1.00	<3.00	2	2
33	LK1FSC03	4/23/2003	0735	1	<2.00	<1.00	<1.00	<3.00	2	2
2004										
14	SS1TWS04	4/26/2004	1250	3	<2.00	<1.00	<1.00	<3.00	5	6
17	SS1ASP04	4/27/2004	0835	2	<2.00	<1.00	<1.00	<3.00	4	5

Supplement 5. Analytical results for quality-assurance/quality-control field-equipment blank samples.—Continued

[ng/L, nanograms per liter; mg/L, milligrams per liter; <, less than; µS/cm, microsiemens per centimeter; ANC, acid neutralizing capacity; °C, degrees Celsius; µg/L, micrograms per liter; U, analyzed for but not detected]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Carbonate, whole-water, titration to pH 8.3, laboratory (mg/L)	Hydroxide, whole-water, titration to pH 10.4, laboratory (mg/L)	Sulfate, dissolved (mg/L)	Chloride, dissolved (mg/L)	Solids, dissolved, sum of constituents (mg/L)	Solids, suspended, residue on evaporation at 105°C (mg/L)
Inorganic properties, major ions, and suspended solids, Continued (North Dakota Department of Health Laboratory, Bismarck, North Dakota)									
2003									
18	SS3WDM03	4/24/2003	0825	<1	<1	1.3	<0.3	12	<5.00
33	LK1FSC03	4/23/2003	0735	<1	<1	1.0	<.3	11	<5.00
2004									
14	SS1TWS04	4/26/2004	1250	<1	<1	0.4	<0.3	13	<5.00
17	SS1ASP04	4/27/2004	0835	<1	<1	<.3	<.3	12	<5.00

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Aluminum, dissolved (µg/L)	Antimony, dissolved (µg/L)	Arsenic, dissolved (µg/L)	Barium, dissolved (µg/L)	Beryllium, dissolved (µg/L)	Boron, dissolved (µg/L)
Trace elements (North Dakota Department of Health Laboratory, Bismarck, North Dakota)									
2003									
18	SS3WDM03	4/24/2003	0825	<50	<1	<1.0	<1.0	<1	U
33	LK1FSC03	4/23/2003	0735	<50	<1	<1.0	<1.0	<1	<50
2004									
14	SS1TWS04	4/26/2004	1250	<50	<1	<1.0	8.1	<1	<50
17	SS1ASP04	4/27/2004	0835	<50	<1	1.3	4.2	<1	<50

Supplement 5. Analytical results for quality-assurance/quality-control field-equipment blank samples.—Continued

[ng/L, nanograms per liter; mg/L, milligrams per liter; <, less than; µS/cm, microsiemens per centimeter; ANC, acid neutralizing capacity; °C, degrees Celsius; µg/L, micrograms per liter; U, analyzed for but not detected]

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Cadmium, dissolved (µg/L)	Chromium, dissolved (µg/L)	Copper, dissolved (µg/L)	Iron, dissolved (µg/L)	Lead, dissolved (µg/L)	Manganese, dissolved (µg/L)
Trace elements, Continued (North Dakota Department of Health Laboratory, Bismarck, North Dakota)									
2003									
18	SS3WDM03	4/24/2003	0825	<1	<1	<1	<10	<1	<10
33	LK1FSC03	4/23/2003	0735	<1	<1	<1	<10	<1	<10
2004									
14	SS1TWS04	4/26/2004	1250	<1	<1	<1	10	<1	<10
17	SS1ASP04	4/27/2004	0835	<1	<1	<1	10	<1	<10

Sampling site number (see figure 1)	Sampling site name	Date of sample collection	Time of sample collection	Nickel, dissolved (µg/L)	Selenium, dissolved (µg/L)	Silver, dissolved (µg/L)	Thallium, dissolved (µg/L)	Zinc, dissolved (µg/L)
Trace elements, Continued (North Dakota Department of Health Laboratory, Bismarck, North Dakota)								
2003								
18	SS3WDM03	4/24/2003	0825	<1	<1	<1	<1.0	<1
33	LK1FSC03	4/23/2003	0735	<1	<1	<1	<1.0	<1
2004								
14	SS1TWS04	4/26/2004	1250	<1	<1	<1	<1.0	1
17	SS1ASP04	4/27/2004	0835	<1	4	<1	<1.0	1

Supplement 6. Analytical results and relative standard deviations for quality-assurance/quality-control replicate samples for sites 1, 19, 26, and 32.

[°C, degrees Celsius]

	Specific conductance, laboratory	Calcium, dissolved	Magnesium, dissolved	Potassium, dissolved	Sodium, dissolved	Acid neutralizing capacity, whole-water, titration to pH 4.5, laboratory	Bicarbonate, whole-water, titration to pH 4.5, laboratory
Number of replicate pairs with complete analytical results for given parameter	4	4	4	4	4	4	4
Median relative standard deviation, in percent, for all replicate pairs	.7	.7	.6	.4	2.0	1.7	1.6

	Carbonate, whole-water, titration to pH 8.3, laboratory	Hydroxide, whole-water, titration to pH 10.4, laboratory	Sulfate, dissolved	Chloride, dissolved	Solids, dissolved, sum of constituents	Solids, suspended, residue on evaporation at 105°C	Aluminum, dissolved
Number of replicate pairs with complete analytical results for given parameter	4	4	4	4	4	4	4
Median relative standard deviation, in percent, for all replicate pairs	0	0	1.7	.8	.9	54.5	46.5

	Antimony, dissolved	Arsenic, dissolved	Barium, dissolved	Beryllium, dissolved	Boron, dissolved	Cadmium, dissolved	Chromium, dissolved
Number of replicate pairs with complete analytical results for given parameter	4	4	4	4	4	4	4
Median relative standard deviation, in percent, for all replicate pairs	0	6.1	7.8	0	6.4	0	0

	Copper, dissolved	Iron, dissolved	Lead, dissolved	Manganese, dissolved	Nickel, dissolved	Selenium, dissolved	Silver, dissolved
Number of replicate pairs with complete analytical results for given parameter	4	4	4	4	4	4	4
Median relative standard deviation, in percent, for all replicate pairs	0	26.2	0	19.6	9.0	0	0

Supplement 6. Analytical results and relative standard deviations for quality-assurance/quality-control replicate samples for sites 1, 19, 26, and 32.—Continued

[°C, degrees Celsius]

	Thallium, dissolved	Zinc, dissolved	Methylmercury, dissolved	Methylmercury, suspended	Methylmercury, whole-water	Total mercury, dissolved	Total mercury, suspended
Number of replicate pairs with complete analytical results for given parameter	4	4	4	3	3	4	4
Median relative standard deviation, in percent, for all replicate pairs	0	14.1	17.4	64.6	7.4	5.1	33.3

	Total mercury, whole-water	Organic carbon, dissolved	Organic carbon, whole-water	Methylmercury, sediment	Total mercury, sediment	Sediment, dry weight	Loss on ignition, sediment, dry weight
Number of replicate pairs with complete analytical results for given parameter	4	4	4	4	4	4	4
Median relative standard deviation, in percent, for all replicate pairs	6.9	.8	1.2	14.1	18.4	7.5	10.8

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