

## MOVEMENT OF ROAD SALT TO A SMALL NEW HAMPSHIRE LAKE

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(Received 10 February 1997; accepted 11 December 1997)

**Abstract.** Runoff of road salt from an interstate highway in New Hampshire has led to contamination of a lake and a stream that flows into the lake, in spite of the construction of a diversion berm to divert road salt runoff out of the lake drainage basin. Chloride concentration in the stream has increased by over an order of magnitude during the 23 yr since the highway was opened, and chloride concentration in the lake has tripled. Road salt moves to the lake primarily via the contaminated stream, which provides 53% of all the chloride to the lake and only 3% of the total streamflow to the lake. The stream receives discharge of salty water from leakage through the diversion berm. Uncontaminated ground water dilutes the stream downstream of the berm. However, reversals of gradient during summer months, likely caused by transpiration from deciduous trees, result in flow of contaminated stream water into the adjacent ground water along the lowest 40-m reach of the stream. This contaminated ground water then discharges into the lake along a 70-m-wide segment of lake shore. Road salt is pervasive in the bedrock between the highway and the lake, but was not detected at all of the wells in the glacial overburden. Of the 500 m of shoreline that could receive discharge of salty ground water directly from the highway, only a 50-m-long segment appears to be contaminated.

**Key words:** cation exchange, ground water, lake contamination, road salt, stream contamination

### 1. Introduction

Contamination of land and water resources from runoff of deicers applied to roadways is common throughout the northern and especially northeastern United States (e.g. D'Itri, 1992; National Research Council, 1991). Many case studies of road-salt contamination of rivers (Peters and Turk, 1981), wetlands (Wilcox, 1986), lakes (Judd, 1970; Bubeck *et al.*, 1971; Cherkauer and Ostenso, 1976; Driscoll *et al.*, 1991), and ground water (Bowser, 1992, Church and Friesz, 1993) have been published. However, for many studies, evidence of the effects of road salt on water supplies is circumstantial (Jones and Jeffrey, 1992). Often this is due to a lack of data prior to application of road salt. The long-term record of chemical data at Mirror Lake, New Hampshire, extending in time both before and after the



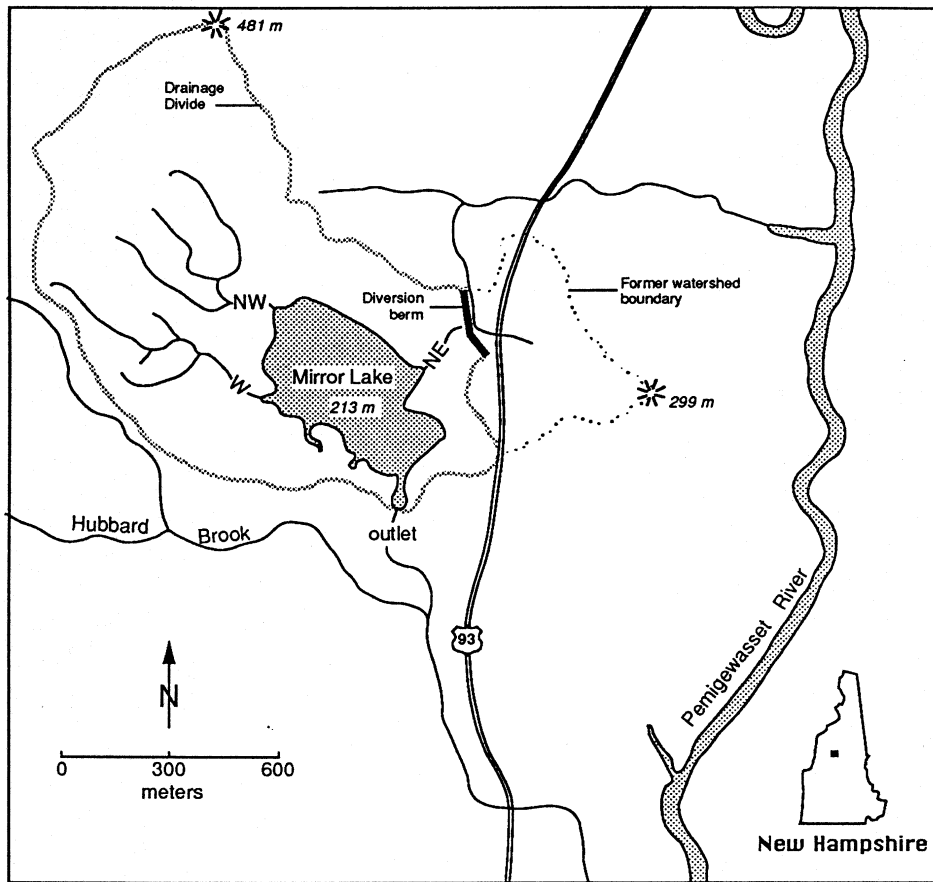


Figure 1. Location, and streams in and adjacent to Mirror Lake watershed.

construction of an interstate highway through the drainage basin, provided a unique opportunity to accurately document the effect of road-salt contamination on the water chemistry of a dilute freshwater lake (Bormann and Likens, 1985). Road-salt contamination of the lake appeared to be caused by direct input of surface drainage via the northeast inlet stream, in part due to a leaky diversion berm, and to contaminated ground water discharge either to the stream or directly to the lake. The relative proportions of these sources were unknown.

Mirror Lake is a small (15 ha, max. depth 11 m), oligotrophic lake in the White Mountains of New Hampshire (Figure 1). During 1969–71, interstate highway I93 was constructed through a portion of the Mirror Lake drainage basin. Because of concerns about contamination of the lake from erosion due to construction activities and road-salt runoff, a diversion berm was constructed to divert surface runoff from the highway to a lowland north of the Mirror Lake drainage basin. This diversion of surface flow reduced the size of the drainage basin of the northeast

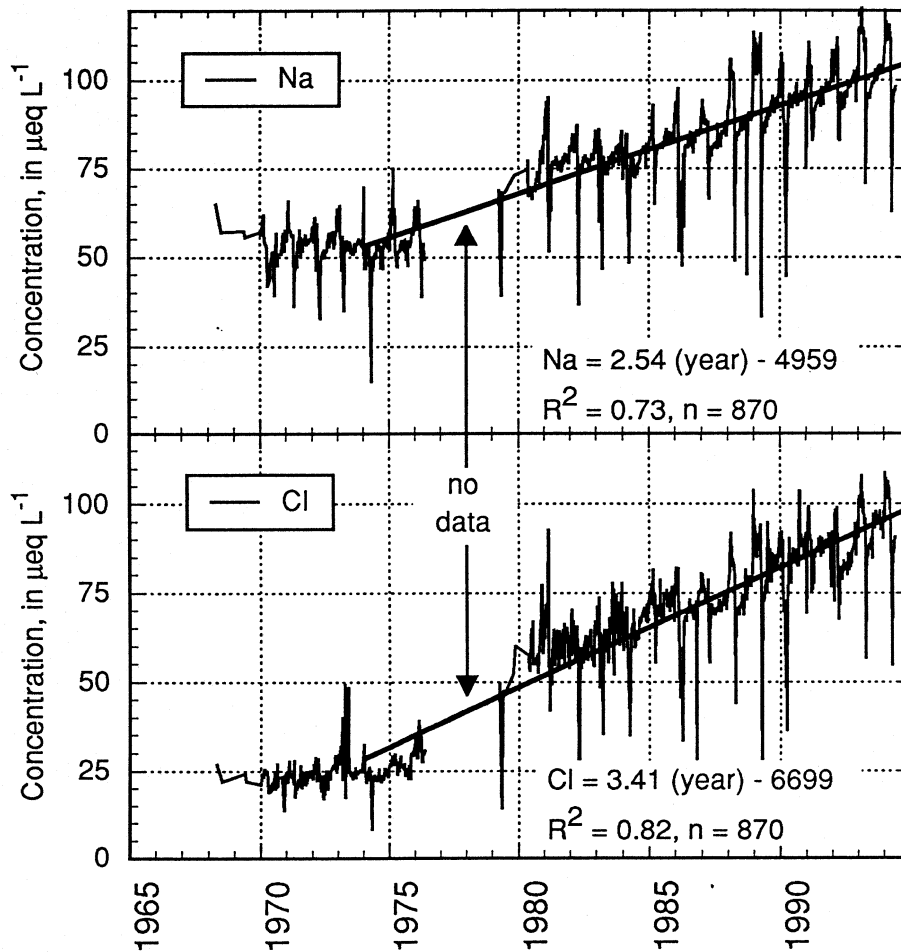


Figure 2. Concentrations of sodium and chloride in Mirror Lake, 1967–1994, in  $\mu\text{eq L}^{-1}$ . Linear regression based on data from the period 1974–94.

inlet stream from 20 to 2.5 ha. In spite of these precautions, concentrations of sodium and chloride in the northeast inlet stream to Mirror Lake have increased by more than an order of magnitude. In Mirror Lake, concentrations of sodium have doubled and concentrations of chloride have more than tripled since about 1973, causing concern about alteration of the ecology of the lake.

From the mid 1960s until the early 1970s, concentrations of sodium in Mirror Lake ranged between 35 and 70  $\mu\text{eq L}^{-1}$  and chloride concentrations ranged between 15 and 30  $\mu\text{eq L}^{-1}$ . Increased concentrations of sodium and chloride first were detected in the lake around 1974–75. By 1980, 9 yr after completion of the interstate highway through the Mirror Lake watershed, concentrations of sodium

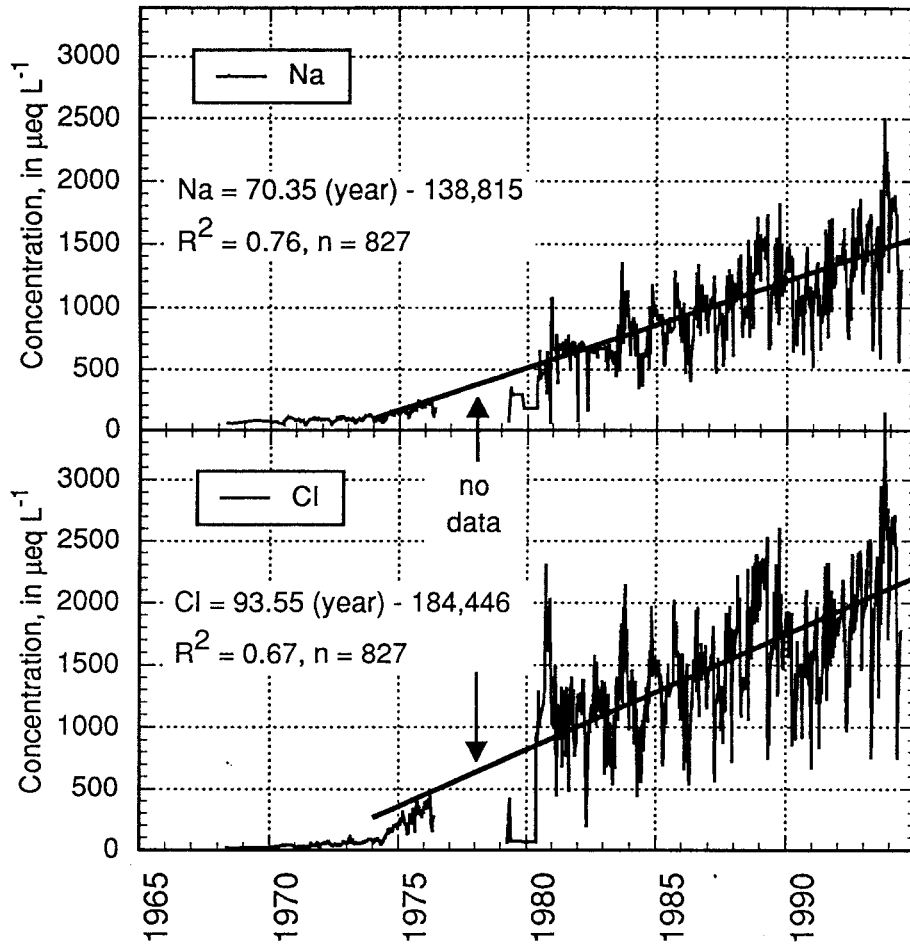


Figure 3. Concentrations of sodium and chloride in the northeast inlet stream, 1967–1994, in  $\mu\text{eq L}^{-1}$ . Linear regression based on data from the period 1974–1994.

had increased by about 40% and chloride in the lake had approximately doubled. Since 1974, a steady increase in concentration of  $2.4 \mu\text{eq L}^{-1} \text{yr}^{-1}$  for sodium and  $3.3 \mu\text{eq L}^{-1} \text{yr}^{-1}$  for chloride has resulted in present-day (1994) concentrations of about  $95 \mu\text{eq L}^{-1}$  sodium and  $85 \mu\text{eq L}^{-1}$  chloride (Figure 2). Present-day concentrations still are quite low, relative to many other studies of lake contamination from road-salt runoff. However, if the rate of road-salt loading to the lake continues to increase, future concentrations could affect the physical and biological processes within the lake, eventually affecting lake circulation, as has happened at numerous other lakes that receive road-salt runoff (Bubeck *et al.*, 1971; Hawkins and Judd, 1972; Cherkauer and Ostenso, 1976).

During the past thirty years, concentrations of sodium and chloride have fluc-

tuated between 20 and 125  $\mu\text{eq L}^{-1}$  in the west and northwest tributaries without any obvious longterm trends. However, at the northeast inlet stream, concentrations of sodium have increased from 100 to 1500  $\mu\text{eq L}^{-1}$  and chloride concentrations have increased from 100 to 2000  $\mu\text{eq L}^{-1}$  from about 1971 to the present (1994) (Figure 3) (Bormann and Likens, 1985; Likens, 1992). Although other sources for sodium and chloride in the Mirror Lake watershed may have contributed to the increased sodium and chloride concentrations in the lake, because of the magnitude and timing of the change it was suspected that road-salt deicers were the primary source of contamination.

### 1.1. PURPOSE AND SCOPE

The pathways by which highway runoff moves from the catchment to the lake needed to be identified to understand why mitigation of road salt runoff still resulted in contamination of the lake, and to allow for better estimates of future concentrations of sodium and chloride in the northeast inlet stream and in the lake. The study also investigated the relative magnitudes of various sources of sodium and chloride to the lake to determine whether increased concentrations of chloride in the lake could be attributed entirely to contributions from the northeast inlet stream, or if other sources of chloride needed to be identified to account for the mass of chloride in the lake. The focus of this investigation was limited to the northeast inlet stream and eastern shoreline of the lake because those areas of the watershed are most susceptible to contamination by movement of road salt from the nearby highway, due to their close proximity to the highway, and because this was where increased sodium and chloride concentrations have been documented. Data specific to this study were collected from late 1992 through August 1994, in conjunction with collection of ongoing long-term monitoring data.

## 2. Physical and climatic setting

The Mirror Lake watershed is located in the northeast section of the Hubbard Brook watershed, which has been the focus of long-term research on the ecology and biogeochemistry of small, forested catchments (Likens and Bormann, 1995; Bormann and Likens, 1979). Hydrologic and limnologic research has occurred within the lake watershed since the early 1960s (Likens, 1985). The Mirror Lake watershed has an area of 85 ha (excluding the lake) and a maximum relief of 268 m. Crystalline bedrock is exposed along parts of the northeast and east shorelines of the lake; bedrock outcrops are present throughout the watershed. Glacial deposits cover most of the watershed, and underlie most of the lake. The glacial deposits generally are a silty, sandy till containing numerous cobbles and boulders. Terrain generally is steep within the watershed, especially on the north and west sides. Till on the east side of Mirror Lake, the area of interest for this study, generally is thin,

ranging from 0 to 11m thick. Three streams flow into Mirror Lake on the west, northwest, and northeast sides of the lake. A dammed outlet on the south side of the lake discharges water to Hubbard brook, which empties into the Pemigewasset River 3 km southeast of the lake (Figure 1).

The northeast inlet stream has an average gradient of 0.04 along the 190 m distance between the diversion berm and the lake, but the gradient steepens along the 50 m segment closest to the lake. A much steeper gradient (about 0.1) exists across the berm between the ponded water east of the berm, and the marshy area directly west of the berm; commonly there is a head difference of 1 m from one side of the berm to the other.

The climate of the Mirror Lake area is humid continental (Likens and Bormann, 1995). Annual precipitation averaged 1400 mm and annual evapotranspiration averaged 525 mm during 1963–1993. Approximately 30% of annual precipitation falls as snow, which generally melts during March and April. Snowmelt contributes to heavy springtime streamflow; about 50% of annual streamflow occurs during March through May. The growing season for the mixed deciduous-coniferous forest in the watershed is from mid May to mid October. Streamflow generally is low during this period (Likens and Bormann, 1995).

### 3. Methods

For this study, two transects, each consisting of two wells and a stream-stage gage, were installed to determine the interaction between ground water and surface water at the northeast stream (Figure 4). Wells were hand augered, 5.1 cm in diameter, contained a 0.5 m long wire-wound PVC screen, and were completed about a meter below the water table. Wells 28 and 30 were completed east of the stream, and wells 29 and 31 were completed west of the stream; all wells were located within 5 m of the stream. A flume (S1 in Figure 4) located at transect 1 provided continuous record of stage and discharge in the stream. This also was the site where long-term discharge and water-chemistry data were collected. At transect 2, a manually-read stage gage (S2) provided stream-elevation data. Stream-stage gages and sampling sites also were established west (downgradient) and east (upgradient) of the diversion berm, designated S3 and S4 respectively (Figure 4). Transect 1 is located approximately 150 m downstream of the berm (20 m upstream of the lake) and transect 2 is located approximately 70 m downstream of the berm (100 m upstream of the lake). Water-level data were recorded weekly; water-table gradients between the northeast stream and adjacent ground water were monitored at these transects from October 1992 to July 1994. Fluxes were estimated using the Darcy equation:  $Q = KIA$ , where  $Q$  = discharge ( $L^3T^{-1}$ ),  $K$  = hydraulic conductivity ( $LT^{-1}$ ),  $I$  = hydraulic gradient (dimensionless) and  $A$  = cross-sectional area through which ground water flows to or from the stream ( $L^2$ ).

Wells 28 and 30, were installed in poorly sorted silty sandy till containing many

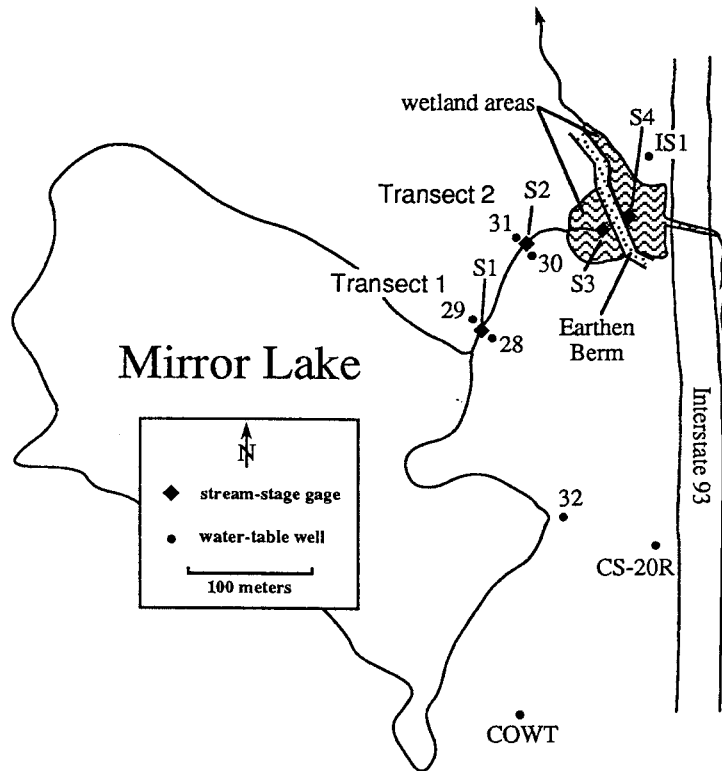


Figure 4. Location of wells and staff gages in study area.

cobbles and boulders; the till at well 28 was particularly silty. Well 31 was installed in till that was less silty than at wells 28 or 30. Well 29 was installed in sand that contained very little silt. Because of low water levels in wells, in-situ hydraulic conductivity was determined by slug test only at well 31, which had a value of  $5 \times 10^{-4} \text{ cm s}^{-1}$ , typical of silty sandy till. Based on observations of geology during well installation, hydraulic conductivity was estimated to be the same for well 30 as for well 31, half an order of magnitude greater at well 29, and an order of magnitude less at well 28.

Three wells were installed adjacent to I93 to obtain information on the vertical gradients within the ground water system between the glacial deposits and the underlying fractured bedrock. Only the water-table well, labeled IS1, is shown in Figure 4. Two of the wells were installed in the glacial deposits at depths of 5.6 and 11.6 m below land surface; each well has a 0.6-m-long well screen. The third well was cased through the unconsolidated glacial deposits and extends as an open hole in the bedrock to 151.8 m below land surface. Additional wells (COWT, 32, CS-20R) were installed to determine ground water gradients in the area between I93 and the eastern side of the lake (Figure 4). Numerous wells completed in bedrock

are located near well COWT, but are not shown in the figure. In addition, numerous other stream gages, wells, rain gages, and other meteorological instruments exist in the Mirror Lake watershed; only those pertinent to this study are shown in Figure 4. All wells, stream-stage gages, and the lake gage were surveyed to a common elevation. Shapiro *et al.* (1995) provide an overview of ground water flow and chemical transport through fractured rock in the watershed.

A hydraulic potentiometer (Winter *et al.*, 1988) was used during July and August 1994 to investigate whether salt-contaminated ground water was discharging into Mirror Lake along the eastern shoreline. This device was used to determine direction of flow between ground water and the lake, and to collect water-chemistry samples from directly beneath the lake bed at distances from 0 to 4 meters from shore. Each hydraulic-head measurement indicated a vertical potential between the point at which the 8-cm-long screen was exposed to the aquifer and the lake. Head measurements relative to lake level were made at three to four distances from shore at each sampling site; with the probe inserted 0.5 m into the lakebed, the smallest vertical hydraulic-head gradient that could be detected was 0.002. The device also was used along the northeast inlet stream to collect hydraulic-head and chemistry data from beneath the streambed.

Water and chemical budgets for Mirror lake were determined to estimate the relative contribution of sodium and chloride from the northeast inlet. Information on methods for determining monthly and annual water and/or chemical budgets for Mirror Lake is presented in Likens (1985), Winter (1984), Winter (1985), and Rosenberry and Winter (1993). Lake chemistry data were collected near the center and deepest portion of the lake.

Precipitation, stream, and lake-water samples were collected weekly for selected chemical components. Samples were collected from wells 28–32 and CS-20R approximately monthly from November 1992 to August 1994. Samples were collected from well COWT intermittently during the study. In-situ temperature, pH, specific conductance, and acid-neutralizing capacity (ANC) also were measured in wells 28–32 during each sampling event. Samples collected with the hydraulic potentiometer system were analyzed for selected major ions, pH and specific conductance. Sodium, calcium, magnesium and potassium were determined by atomic adsorption spectrophotometry. Chloride, sulfate, nitrate, phosphorus and ammonium were determined by ion chromatography or by automated spectrophotometric methods. ANC was determined by titration with hydrochloric acid using a granplot analysis of automated potentiometric pH data, and pH was determined by potentiometry with glass electrodes calibrated at pH 7 and 4 (Wetzel and Likens, 1991; Likens and Bormann, 1995).



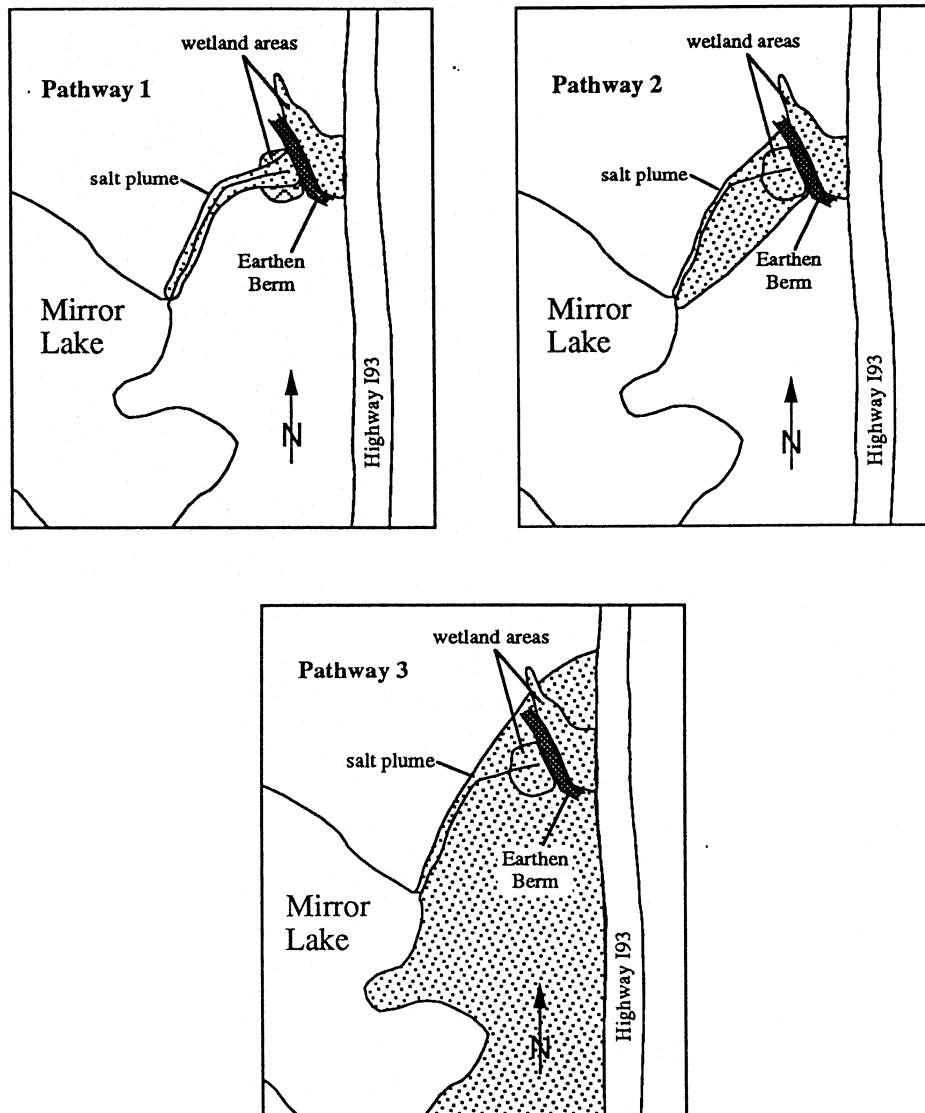


Figure 5. Potential pathways for movement of road salt between I93 and Mirror Lake.

#### 4. Results and Discussion

##### 4.1. PATHWAYS FOR CONCENTRATION

Three potential pathways for movement of road salt to Mirror Lake are shown in Figure 5. (Pathway 1) A narrow zone of salt-contaminated water seeps through the diversion berm and discharges directly to the present-day headwaters of the northeast stream. Ground-water gradients are assumed to be toward the stream,

which would restrict salt contamination to a narrow region along the stream. Salt travels via the stream to the lake both in the stream and also in the narrow, shallow, hyporheic zone directly beneath and adjacent to the stream. (Pathway 2). Water that has seeped through the berm discharges both to the headwaters of the stream as well as to ground water on either side of the stream, directly downgradient of the berm. Salt-contaminated ground water flows toward the lake, but lateral ground water gradients toward the stream cause the plume to merge with the stream before flowing as stream water and hyporheic water to the lake. Because ground water wells located north of the lake have water levels much higher than in the vicinity of the northeast stream, and because the stream is incised relative to the hillside directly west of the stream, it is assumed that ground water gradients toward the stream on the west of the stream prevent a salt plume from spreading west of the stream. (Pathway 3) Salt contamination is not restricted to the northeast stream headwaters region. A broad region of salt-contaminated ground water moves from the interstate highway toward the entire eastern shoreline of Mirror Lake. As with the second pathway, an assumed gradient toward the stream on its west side prevents movement of a plume to the west of the stream. The third pathway provides the greatest potential for future large increases in concentrations of sodium and chloride in the lake.

#### 4.2. HYDROLOGY AND CHEMISTRY OF THE NORTHEAST STREAM

Streamflow in the northeast stream normally reaches a maximum in April, declines throughout the summer to a minimum in July or August, then rises to a smaller peak in the fall before declining again during winter (Figure 6). From June through October, the northeast stream often flows only following rain events. Chloride concentration typically is diluted during spring runoff, but concentration is little affected by increased streamflow in the fall. The mass of chloride that enters Mirror Lake from the northeast stream has a bimodal peak; the greatest mass enters during April, coincident with the streamflow peak, and a slightly smaller peak also occurs during October and November (Figure 6). A similar bimodal pattern for flux of calcium, sulfate and silicate from all streams into Mirror Lake was reported in Likens *et al.* (1985).

Chloride concentrations in the northeast stream usually decreased with distance from I-93 (Figure 7A, Table I), Median chloride concentrations were 4202  $\mu\text{eq L}^{-1}$  at S4, 3308  $\mu\text{eq L}^{-1}$  at S3, 2552  $\mu\text{eq L}^{-1}$  at S2 and 1472  $\mu\text{eq L}^{-1}$  at S1. Occasionally chloride concentration increased downstream from the berm between sites S3 and S2. For example, from March through June 1993, chloride concentration at site S2 was higher than at site S3 (Figure 7A). Also, occasionally chloride concentrations were lower in the ponded water on the highway side (east) of the berm, an example of which was from December 1993 through April 1994, when chloride concentration on the east side of the berm at site S4 was lower than at both sites S3 and S2 (Figure 7A).

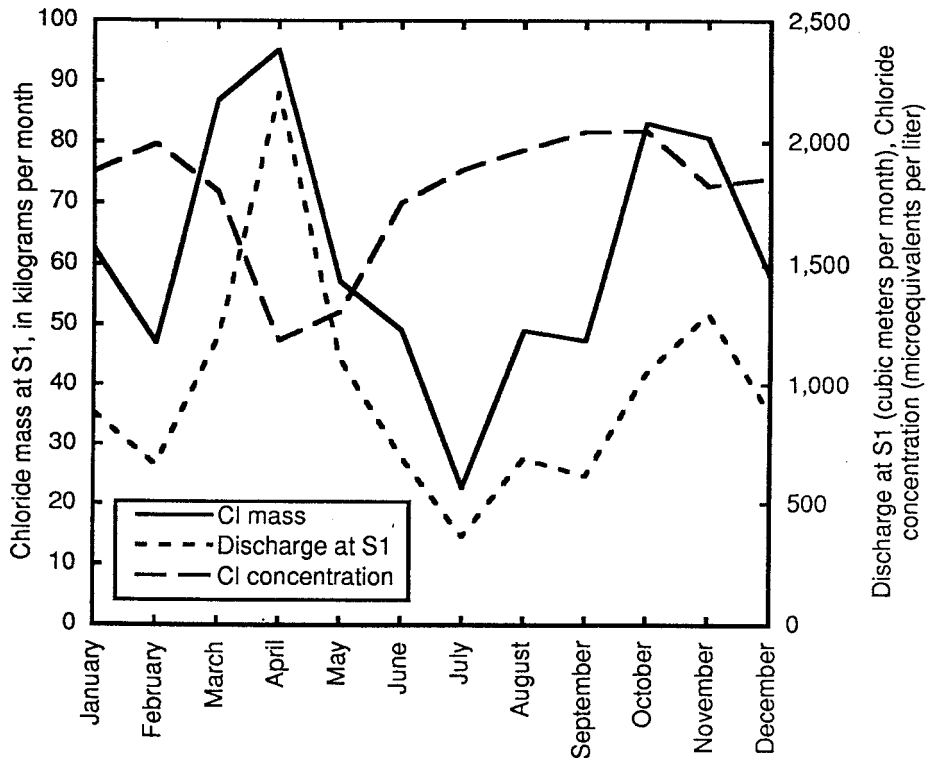


Figure 6. Monthly flow ( $\text{m}^3 \text{mo}^{-1}$ ), chloride concentration ( $\mu\text{eq L}^{-1}$ ), and chloride mass ( $\text{kg mo}^{-1}$ ) in the northeast stream (medians of monthly values from 1988–1993).

Assuming that chloride is conserved, declining concentration in the stream indicates that uncontaminated ground water discharged into the stream and diluted the stream water. The average dilution of chloride between sites S3 and S1 indicates that streamflow increased in volume by about 35% over a stream reach of approximately 150 m. Estimated ground water fluxes, using average gradients at the two transects and a hydraulic conductivity ranging from  $5 \times 10^{-4} \text{ cm s}^{-1}$  to  $1 \times 10^{-3} \text{ cm s}^{-1}$ , indicated a similar increase in streamflow, between 7 and 33%. Estimated fluxes ranged from 65 to  $330 \text{ m}^3 \text{mo}^{-1}$  between sites S3 and S1.

Water-chemistry samples from wells adjacent to the stream indicated that ground water in the vicinity of the stream is not contaminated with road salt, except in the vicinity of well 29 (Figure 7B). Chloride concentrations at well 29 were approximately an order of magnitude greater than concentrations at wells 28, 30 and 31, which had chloride concentrations similar to uncontaminated ground water west of Mirror Lake. However, concentrations at well 29 still were much lower than in the stream, indicating that some of the water flowing to the well did not originate from the stream.

TABLE I

Median values for selected chemical constituents (and ratios relative to sodium) from 11 samples collected from the northeast inlet stream, January 1991 – August 1994, in  $\mu\text{eq L}^{-1}$

Site	Ca	Mg	K	Na	H	SO <sub>4</sub>	NO <sub>3</sub>	Cl	ANC
S1	514	160	33	1394	3	143	4	1472	71
S2	656	206	46	1881	4	143	4	2552	183
S3	1794	568	92	1960	1	143	2	3308	1400
S4	781	225	98	3678	2	143	4	4202	1263

Site	Na/Ca	Na/Mg	Na/K	Na/Cl
S1	2.7	8.7	42.2	0.9
S2	2.9	9.1	40.9	0.7
S3	1.1	3.5	21.3	0.6
S4	4.7	16.3	37.5	0.9

Notes:

1. Ammonium and ortho-phosphate were measured only at site S1 and were assumed to be negligible at all other sites.
2. Obvious rusty flocculate in samples from sites S3 and S4 indicate missing cation may be  $\text{Fe}^{+3}$ .

Water-level data collected at the two transects perpendicular to the northeast stream indicated that water-table gradients were not always toward the stream as had been expected, and as the water-chemistry data indicated. During fall through spring, gradients typically were toward the stream at both transects (Figure 8). However, during early to mid summer, gradients reversed as the water table dropped below the stream level at both transects. Gradients were away from the stream during most of the summer, until cessation of transpiration likely allowed fall recharge to raise the water table above the stream stage. Maximum flow velocities between the stream and ground water at wells 28-31, assuming a porosity of 0.2 (based on the poorly sorted till adjacent to the stream) and using the maximum gradients recorded between the wells and the stream, ranged from about  $0.02 \text{ m day}^{-1}$  at well 28 to about  $0.5 \text{ m day}^{-1}$  at well 29. Gradients at these wells frequently were much smaller than the maximum recorded gradients; in addition, gradients reversed seasonally. Therefore, net movement of salt-contaminated ground water in the vicinity of the stream was significant only in the vicinity of well 29.

#### 4.3. CONTAMINATION OF GROUND WATER FROM THE NORTHEAST INLET

Discrete measurements of water levels at transect 1, along with large levels of chloride at well 29, indicated that the lower reach of the stream was losing water, a condition not expected prior to this study. The steeply sloping hillside to the

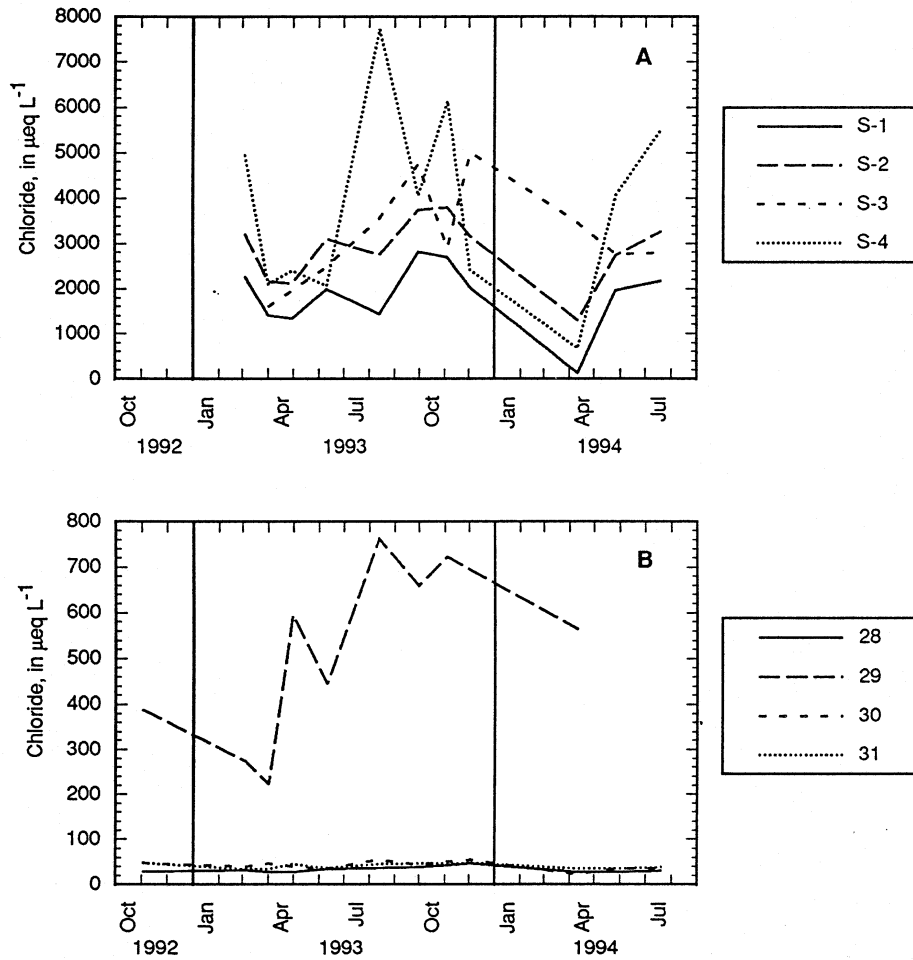


Figure 7. Concentration of chloride ( $\mu\text{eq L}^{-1}$ ): (A) at sites S1-S4, (B) at wells 28-31.

west of the stream, together with ground water levels north of the lake that were 30 m higher than the lake, had led to the assumption that gradients toward the stream from the west would be steep. Wells 28 and 29 were instrumented with continuous water-level-monitoring equipment from mid July to early October 1994 to determine if diurnal ground water fluctuations would show evidence of transpiration, which could remove ground water and create a gradient from the stream to ground water. During much of the period of record, water-level fluctuations in the wells showed little evidence of diurnal fluctuations. Water levels usually were rising or falling rapidly in response to individual rainfall events, which would mask much smaller fluctuations due to transpiration. However, an extended dry period during the first two weeks of August provided evidence that evapotranspiration

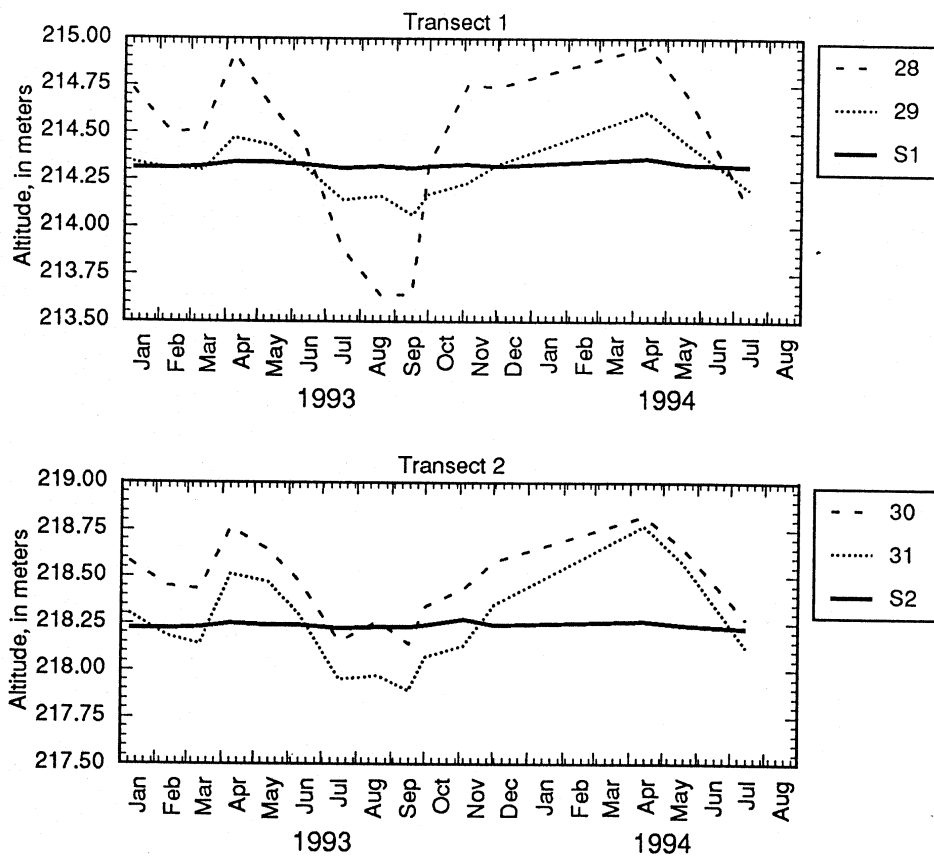


Figure 8. Stream and ground-water-head altitudes at two transects across the northeast stream, January 1993 through August 1994, in m.

was removing ground water (Figure 9). At well 29, where the geologic deposits are relatively permeable, response to evapotranspiration was subtle but immediate. At well 28, where the till conducts water much less readily than the sand at well 29, water-level fluctuations were larger, but they were delayed. At well 29, on all but one day during August 2–12, the water-level declined in early afternoon during times of maximum evapotranspirational withdrawals. On August 9, 11, 12, and 13, the water level recovered slightly during the nighttime and early morning hours. Light showers on August 2, 5, and 9 may have reduced slightly the water-table decline by reducing transpiration. These data indicate that evapotranspiration was responsible for drawing ground water down to below stream level at transect 1. This process probably occurred at transect 2 as well, but hourly data were not collected from that site.

Exchange of stream water with water in the hyporheic zone adjacent to the stream (White, 1993) may also have contributed to contamination of ground water

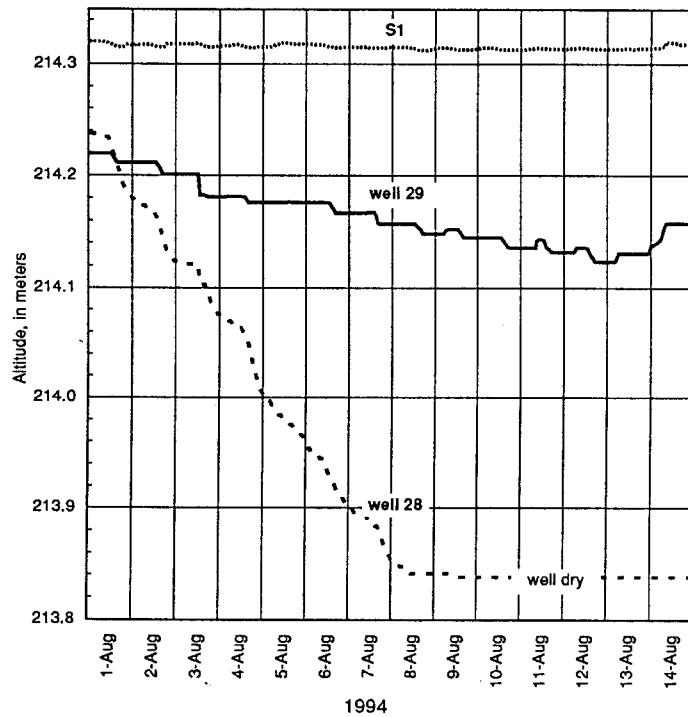


Figure 9. Hourly fluctuation of stream stage and ground water head at site S1, well 28, and well 29, August 1–14, 1994, in m.

in the vicinity of well 29. The pool and riffle nature of the northeast inlet stream may have created short stream-reach segments where the gradient was from pooled stream water to ground water, even when gradients at transects 1 and 2 were toward the stream. In such an event, once stream water with large concentrations of salt flowed into the ground water system, that salty water could travel through the ground water system downvalley toward the lake essentially parallel to the stream, contaminating ground water downstream even in locations where the downstream ground water was at a higher head than the stream water at that downstream location. Evidence that this process was occurring is presented in Figures 7B and 8. During April 1993, the concentration of chloride increased sharply at well 29 (Figure 7B) while at the same time the head at well 29 was higher than the stream stage at S1 (Figure 8). Since stream water directly adjacent to well 29 could not flow to well 29, it is possible that stream water entered the ground water system somewhere upstream, and flowed downvalley to well 29, causing the increase in chloride concentration observed during April 1993.

#### 4.4. EVIDENCE OF SALT CONTAMINATION IN THE NORTHEAST INLET STREAMBED

Vertical hydraulic-head gradients were measured with the hydraulic potentiometer during late July 1994 beneath the streambed, and chemistry samples were collected at the same time. At two sampling locations within 10 m of transect 2, vertical gradients were small (0.01, 0.02) and indicated seepage from the stream to ground water. A measurement made in a small pool in the stream halfway between S2 and S3 indicated a large gradient (0.13) from the stream to ground water. In the vicinity of the berm gradients also were large; west of (below) the berm the gradient was 0.24 from ground water to the stream, and the gradient was 0.07 from ponded water to ground water east of the berm.

Large chloride concentrations were measured in ground water to depths of 0.4 and 0.6 m directly beneath the stream, which was as far as the probe was inserted. Chloride concentration beneath the streambed was largest directly east of the berm ( $5590 \mu\text{eq L}^{-1}$ ) and directly west of the berm ( $5080 \mu\text{eq L}^{-1}$ ). Sub-streambed chloride concentrations ranged between 2300 and  $2500 \mu\text{eq L}^{-1}$  along a 50-m long reach centered on transect 2. Even at transect 1, where the hydraulic gradient was very small, chloride concentration beneath the streambed still was large ( $1780 \mu\text{eq L}^{-1}$ ) relative to uncontaminated ground water. It is likely that in the fall, when reduced evapotranspiration and increased recharge cause a reversal in the hydraulic gradient between the stream and contiguous ground water, some of the salt beneath the streambed is flushed into the stream, and contributes to the increase in chloride concentration in the stream during October and November.

#### 4.5. DISCHARGE OF SALT TO THE NORTHEAST INLET HEADWATERS AREA

Water chemistry data (Table I) collected at the four sites along the northeast inlet (locations shown in Figure 4) indicate that ground water may have discharged to the northeast stream via three different flow paths. (1) Water may have flowed through the berm, from the pond east of the berm to the headwaters area west of the berm. This flow path is indicated by the difference in water chemistry between sites S4 and S3. The median water chemistry of samples collected at site S3 indicated 130% more calcium, 152% more magnesium, and 47% less sodium than samples collected at site S4. The change in the proportion of base cations indicates that significant cation exchange occurred in the berm material, perhaps due to the existence of many freshly exposed weathering surfaces created during the emplacement of berm fill. (2) Between sites S3 and S2, sodium concentrations in the stream usually changed only slightly while calcium and magnesium concentrations usually decreased, changing considerably the proportions of base cations at site S2 to ratios similar to the water ponded east of the berm at site S4 (Table I). This indicates a second possible ground water flow path; water from the pond east of the berm flowed beneath the berm through undisturbed glacial and streambed deposits and discharged farther downstream, between sites S3 and



S2. Water that flowed via this flow path contained sodium and chloride and little else, did not undergo significant cation exchange, and when mixed with the stream water, resulted in the large decrease in calcium and magnesium concentration in the stream at site S2 relative to site S3. This flow path also is a possibility because it is likely that some water could flow beneath the berm due to the potentially higher hydraulic conductivity of the streambed material beneath the berm relative to the hydraulic conductivity of the berm material. Another explanation for the change in stream water chemistry between sites S3 and S2 is the 'valence dilution effect' associated with cation exchange reactions (Bohn *et al.*, 1985). At elevated concentrations of salt, the ratio of monovalent to divalent cations is increased on the soil/sediment surface. With dilution of the salt between sites S3 and S2, divalent cations are preferentially retained over monovalent cations on the soil-exchange complex, reducing the concentrations of calcium and magnesium in the stream water relative to sodium. (3) Since chloride concentration decreased from S3 to S2, and from S2 to S1, a third flow path could have been uncontaminated ground water that discharged from areas of higher head contiguous to the stream. This flow path likely occurred along most of the stream reach between the berm and the lake.

Prior to this study, there were concerns that cation exchange in the northeast-inlet subcatchment was leaching calcium from the soils in the vicinity of the stream. This study indicates that the bulk of cation exchange takes place within the berm and along a short reach of the stream upstream of site S2. Downstream of site S2, the data indicate little cation exchange takes place; all cations decrease nearly uniformly in response to dilution of the stream from discharge of uncontaminated ground water. During the first five years following completion of I93, ratios of sodium to chloride diminished from 2–3 (similar to ratios from the pristine watershed at nearby Hubbard Brook (Likens and Bormann, 1995)) to less than 1. Since 1980, ratios have steadily increased from an annual mean of 0.4 in 1980 to 0.7 in 1993 (significant at  $p < 0.01$ ), indicating that cation exchange may be diminishing with time. It is possible that the cation exchange in the berm is decreasing with time due to a limited supply of base cations (calcium, magnesium, potassium) in the berm material that is being depleted. Glacial deposits in the local area, from which the berm was constructed, typically contain only 0.2 to 17% calcium by weight; the average is between 1 and 2% (Billings and Wilson, 1965).

#### 4.6. HYDROLOGY AND CHEMISTRY OF GROUND WATER BETWEEN MIRROR LAKE AND I93

Water-level measurements at wells located between Mirror Lake and I93 indicate that ground water flows from the vicinity of I93 to the lake along most of the eastern shoreline (Figure 10). Springs seeping out of the roadcut east of I93, at an elevation much higher than the wells west of the highway, indicate that the gradient across the highway also is toward the lake. Water-table gradients are quite steep, typically 0.04 between the interstate wells and the northeast stream outlet, and 0.1

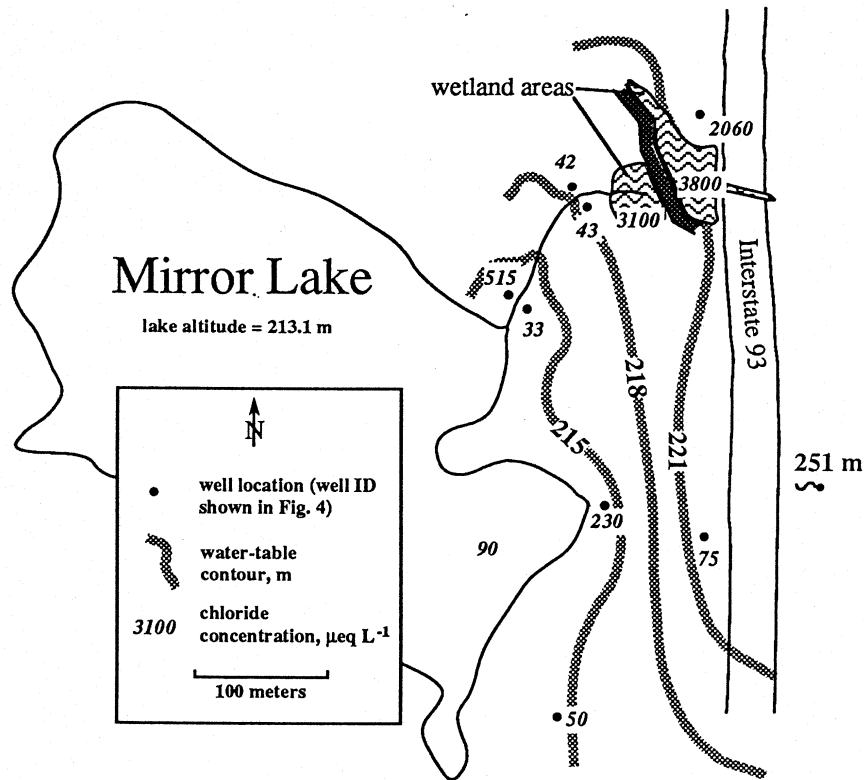


Figure 10. Water-table altitude (m) and chloride concentration ( $\mu\text{eq L}^{-1}$ ) east of Mirror Lake, averaged over study period.

between wells CS-20R and 32. Between the spring east of I93 and well CS-20R the gradient is especially steep and approaches 0.3. Assuming a hydraulic conductivity of  $1 \times 10^{-4} \text{ cm s}^{-1}$  (Rosenberry and Winter, 1993) and a porosity of 0.2, ground water velocity in the area is between 6 and 16 m per year. If salt-contaminated ground water were flowing toward the lake it would take about 7 to 18 yr to reach the easternmost shoreline of Mirror Lake, assuming that flow was restricted to the glacial deposits. If flow were in the fractured bedrock, arrival times could be much shorter or much longer, depending on the number, orientation, and connectivity of fractures in the underlying bedrock.

A lack of wells east of well COWT precludes a determination of hydraulic gradients along the southernmost part of the eastern shoreline of the lake. However, surface topography, and the presence of Hubbard Brook and the Pemigewasset River to the south and east, suggest that flow is either to the south or southeast, which would prevent salt-contaminated ground water from reaching the lake along the southern part of the eastern shoreline.

Median concentrations of chloride for water from selected wells and wetlands are shown in Figure 10. Values of chloride larger than the typical  $50\text{--}70\ \mu\text{eq L}^{-1}$  were found at most wells. Above-normal values were not detected at well COWT, which is a shallow water-table well completed in glacial deposits. However, water samples collected from deeper boreholes in the fractured bedrock beneath well COWT had chloride values between  $110$  and  $1300\ \mu\text{eq L}^{-1}$ . The chloride concentration indicated in Figure 10 for water collected from well IS1 is from a sample collected from an isolated interval of bedrock at a depth of  $17.4$  m below land surface. Samples were collected from three additional isolated intervals in this bedrock borehole, at depths of  $112$ ,  $123$ , and  $143$  m below land surface. Chloride values for these intervals were  $1610$ ,  $1520$ , and  $70\ \mu\text{eq L}^{-1}$  respectively, indicating that road-salt contamination of ground water has penetrated at least  $123$  m into the bedrock beneath the highway.

It appears that road salt has contaminated the ground water system at several locations between I93 and the lake, based on above normal chloride concentrations at wells CS-20R, 32 and wells completed in the bedrock beneath well COWT. However, since chloride concentration at well CS-20R is only slightly larger than uncontaminated ground water in the area (typically  $50\text{--}70\ \mu\text{eq L}^{-1}$ ), salt may not have contaminated all ground water in the glacial deposits between I93 and the lake. It also is possible that above normal concentrations of chloride in the vicinity of well 32 was due not to road salt, but to leachate from a small septic-system drainfield. However, other data do not support the septic-system source for ground water near well 32. Septic leachate usually exhibits larger values for ammonium, phosphorus, and nitrate, and concentrations of all three constituents were small or undetectable at well 32. Hydraulic heads in the bedrock near I93 are much higher than heads near and in Mirror Lake, and it is possible that water contaminated with road salt could move through a network of interconnected fractures in the bedrock, and discharge to the glacial deposits in the vicinity of well 32, where glacial deposits are only a few meters thick. If this were happening, larger concentrations of sodium and chloride would be present near well 32, but not pervasive in the glacial deposits between well 32 and I93.

#### 4.7. HYDROLOGY AND CHEMISTRY ALONG THE EASTERN SHORELINE OF MIRROR LAKE

Directions of seepage along the eastern shoreline of Mirror Lake, as measured by the hydraulic potentiomanometer, are shown in Figure 11. Each arrow in Figure 11 represents one or more measurements made at each sampling transect; a total of 66 sampling locations are presented by the arrows shown along the eastern and northeastern shoreline of Mirror Lake. At most sampling transects, measurements were made at 3 to 4 distances from shore. An attempt was made to insert the probe consistently to  $0.5$  m below the lakebed, but at some locations a shallower insertion was required due to the presence of rocks and boulders. At some of the sampling

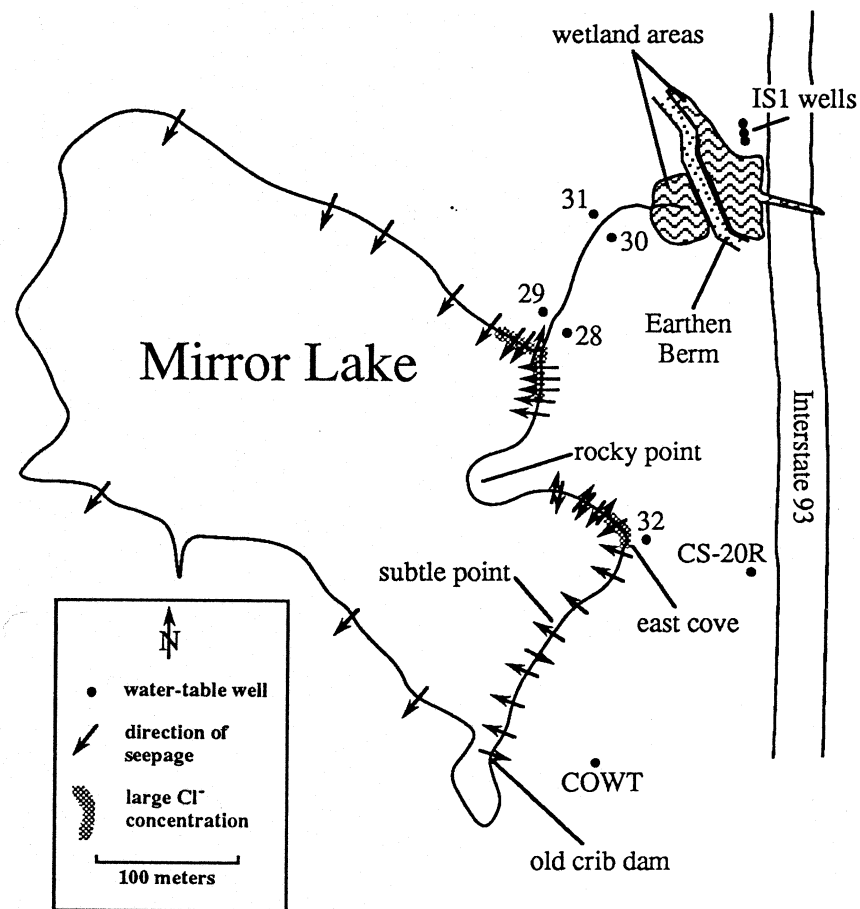


Figure 11. Primary direction of flow, and reaches of elevated sodium and chloride concentrations, along the eastern and northeastern shoreline of Mirror Lake, summer 1994.

transects, seepage direction changed with distance from shore. These small-scale reversals in seepage often are induced by transpiration from near-shore plants. In these situations, the direction that was indicated by the majority of sampling locations was the direction indicated by the arrow on the map. Also, some of the sampling locations were visited two to three times during July and August 1994. In cases where the seepage direction was consistent throughout all visits, only one arrow is indicated for that location, but for cases where the direction reversed from one visit to the next, two arrows, spaced very closely together but pointing in opposite directions, are shown on the map.

Vertical hydraulic-head gradients beneath the lakebed generally were small along the Mirror Lake shoreline between well 32 and the no-longer-functional old crib dam near the outlet of the lake (Figure 11). Many measurements were near the

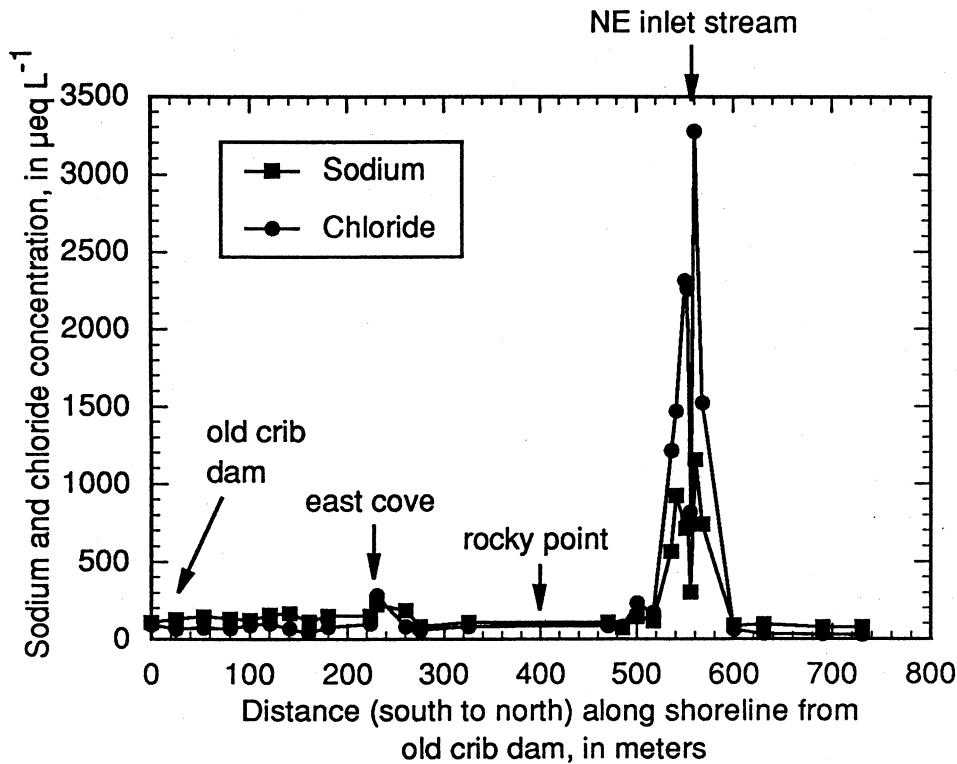


Figure 12. Concentrations of sodium and chloride along eastern and northeastern shoreline of Mirror Lake, summer 1994, in  $\mu\text{eq L}^{-1}$ .

detection limit of the probe (about 0.002). In the vicinity of the old crib dam, gradients were large (0.01 to 0.03) and they uniformly indicated seepage from the lake to ground water. However, along most of the rest of the eastern shoreline of Mirror Lake, gradients indicated seepage from ground water to the lake. Along the section of shoreline between well 32 and rocky point, seepage direction was variable and gradients were small. North of rocky point gradients were larger (0.003 to 0.02), and northwest of the northeast stream inlet gradients were especially large (0.02 to 0.035). Data from all transects in this shoreline reach, except one located at the stream inlet, indicated seepage into the lake; gradients at the transect near the inlet that indicated flow in the opposite direction were questionable because they were so small.

The hydraulic potentiometer also was used to determine gradients and collect water-chemistry samples at a few other shoreline locations along the west and south shorelines. Seepage directions, as indicated in Figure 11, were consistent with data collected in the late 1980s by Asbury (1990).

Median sodium and chloride concentrations of samples collected with the potentiometer are plotted with distance along the shoreline in Figure 12, be-

ginning with 0 at the old crib dam and ending with 730 m along the midpoint of the northern shoreline of the lake. Two areas of above-normal sodium and chloride concentration are indicated in the plot, as well as on Figure 11; one a 50-m shoreline segment in the east cove (distance 220–270), and the other a 70-m shoreline segment bracketing the northeast stream inlet (distance 500–570). At the east-cove shoreline segment, sodium concentration is about 1.7 times the concentrations on either side of the segment, and chloride concentration is about 3.7 times larger than concentrations on either side of the segment. At the contaminated segment near the northeast stream inlet, sodium concentrations are more than 10 times the concentrations adjacent to the segment, and chloride concentrations are more than 40 times concentrations adjacent to the segment.

Average sodium and chloride concentrations between the old crib dam and the east cove were 1.5 and 1.9 times as large as along the northern shoreline directly west of the contaminated segment near the northeast inlet. Sodium and chloride concentrations along the uncontaminated northern shoreline are similar to concentrations in other uncontaminated ground water in the western part of the Mirror Lake watershed. It is possible that the shoreline segment north of the old crib dam has been contaminated by septic systems from seasonal cabins located along that shoreline. However, as mentioned earlier, these systems are small and seasonal, and concentrations of ammonium, phosphorus and nitrate along that segment of shoreline either are very low or undetectable, indicating that septic leachate is not the cause of these slightly greater-than-normal sodium and chloride values. These concentrations also are approximately equal to present-day sodium and chloride concentrations in Mirror Lake. Frequent reversals in direction of seepage along this segment of shoreline may have contributed to sodium and chloride concentrations in the near-shore porous media that are similar to concentrations in the lake.

#### 4.8. MASS-BALANCE APPROACH

The sector of I93 that includes the Mirror Lake watershed is the most heavily salted sector of the highway in New Hampshire (State of New Hampshire written communication, 1995). During the five-winter period from 1986 to 1991, application of sodium-chloride salt varied from 23.2 to 33.7 metric tons per lane mile (State of New Hampshire written communication, 1995). The average annual application (1986–1991) to the section of highway that includes the Mirror Lake watershed was 44.6 metric tons of salt, about 27 metric tons of which was chloride. The annual flux of chloride past site S1 during 1988–1993, based on stream discharge measurements and weekly chloride concentration measurements, was about 0.7 metric ton. Using chloride as an indicator of salt flux, less than three percent of the average annual application of road salt to the Mirror Lake watershed has reached Mirror Lake via the northeast inlet. The rest of the salt either was diverted as surface flow past the berm and out of the watershed, is transferred to ground water, or

TABLE II

Total annual mass of chloride for components of Mirror Lake chloride budget, 1989-1993, in  $\text{kg yr}^{-1}$

		1989	1990	1991	1992	1993	Ave.
	Outlet	673	406	728	755	803	673
	Ground water	838	330	903	890	973	787
Fluxes out	Total out	1511	736	1631	1634	1776	1460
	West inlet	325	619	383	378	237	388
	Northwest inlet	302	428	286	267	237	304
	Northeast inlet	677	954	758	750	708	769
	Precipitation	43	42	23	43	62	43
	Ground water	35	36	35	35	35	35
Fluxes in	Total in	1382	2079	1485	1473	1279	1539
	Budget balance	-129	1343	-146	-172	-497	79

stored in the soil. These data indicate that the berm actually was quite effective at preventing salt from reaching Mirror Lake.

A chloride budget for Mirror Lake was calculated for 1989–1993 to determine chloride input from the northeast stream relative to other sources of chloride to the lake. Weekly data were used to calculate stream fluxes and atmospheric fluxes, and monthly data were used to calculate ground water fluxes. Assumptions were that the lake was uniformly mixed regarding chloride concentration, and that the concentration of chloride that seeped into the ground water and/or that flowed out of the lake via the outlet stream was the same as the concentration in the lake. Dry deposition also was considered to be insignificant based on comparisons of chloride concentration in precipitation collected within the Mirror Lake basin and farther west in the Hubbard Brook valley.

Annual summaries of the results are shown in Table II. The 5-yr average increase in chloride in the lake, based on chloride fluxes to and from the lake, is  $79 \text{ kg yr}^{-1}$ . As a comparison, the 5-yr increase in mass in the lake, based on the change in lake concentration multiplied by the lake volume, is  $72 \text{ kg yr}^{-1}$ . The long-term increase in chloride in the lake, based on a best fit of lake chloride concentration data for the period 1974–1993 and using an average lake volume of  $8.62 \times 10^8 \text{ l}$  (Figure 2), was  $104 \text{ kg yr}^{-1}$ . (For the period 1980–1994, the long-term increase in chloride in the lake was  $82 \text{ kg yr}^{-1}$ ). While the 5-yr-average data compare quite well with loading based on change in measured concentrations of chloride in the lake, for four of the five years chloride budgets indicated a loss of chloride. These

budget results could indicate that a source of chloride exists for the lake that we have not yet observed or measured.

Budget data indicate that the northeast stream provides half of all chloride inputs to the lake. While streamflow from the northeast stream accounts for only 3% of all streamflow to the lake, chloride input from the northeast stream accounts for 53% of all inputs from the streams and 50% of all inputs to the lake when precipitation and ground water also are included.

## 5. Future Salt Loading to the Northeast Stream and Mirror Lake

Since 1974, when increases in sodium and chloride first were detected, the concentration of sodium in the northeast inlet increased at an average annual rate of  $70 \mu\text{eq L}^{-1} \text{yr}^{-1}$  and chloride increased at an average annual rate of  $94 \mu\text{eq L}^{-1} \text{yr}^{-1}$ , based on simple linear regression (Figure 3). If the concentration of sodium and chloride were to continue to increase at the same rates, average annual concentration of sodium could be expected to be near  $1900 \mu\text{eq L}^{-1}$  by the yr 2000 and  $3600 \mu\text{eq L}^{-1}$  by the yr 2025. Average annual concentration of chloride could be expected to be near  $2700 \mu\text{eq L}^{-1}$  by the yr 2000 and  $5000 \mu\text{eq L}^{-1}$  by the yr 2025.

Eventually, concentrations of sodium and chloride in the northeast inlet will begin to stabilize as pore spaces in the soil adjacent to and beneath the headwaters region of the northeast inlet become saturated with salt-laden ground water. There is some indication that the increase in concentration of sodium and chloride in the northeast inlet is abating slightly, 23 yr after salt loading began at I93. Linear regressions of sodium and chloride concentrations in the northeast stream, based on 1974–1984 data, indicate an annual increase of  $64 \mu\text{eq L}^{-1}$  for sodium and  $180 \mu\text{eq L}^{-1}$  for chloride. Linear regressions based on 1985–1994 data indicate annual increases of  $73 \mu\text{eq L}^{-1}$  for sodium, but only  $87 \mu\text{eq L}^{-1}$  for chloride. The larger annual increase of sodium during the latter period indicates that cation exchange sites are being depleted in the northeast headwaters region, decreasing the capability of sodium to be exchanged primarily with calcium, but the smaller annual increase of chloride for the latter period indicates that the concentration of salt in the soil and ground water near the headwaters area is closer to equilibrium with the rate of application of road salt to highway I93.

If the trend of increasing sodium and chloride concentration in the northeast inlet were to continue, the trend of increasing sodium and chloride concentration in Mirror Lake would continue also. Linear regression of data collected from the lake from 1974–1993 indicates an average annual rate of increase of  $2.5 \mu\text{eq L}^{-1} \text{yr}^{-1}$  for sodium and  $3.4 \mu\text{eq L}^{-1} \text{yr}^{-1}$  for chloride (Figure 2). A continued rate of increase would result in a concentration of  $118 \mu\text{eq L}^{-1}$  for sodium and  $116 \mu\text{eq L}^{-1}$  for chloride by the yr 2000, and a concentration of  $181 \mu\text{eq L}^{-1}$  for sodium and  $201 \mu\text{eq L}^{-1}$  for chloride by the yr 2025. By the yr 2025 the concentration of



sodium would be triple the concentration in the lake prior to construction of I93, and the concentration of chloride would have increased 8 fold.

The rate of increase in concentration of salt in Mirror Lake may be diminishing also, as in the northeast stream water, but evidence for a smaller annual increase in concentration in the lake during the past 10 yr is more subtle than in the northeast stream water. The slope of the linear-regression lines through the data collected from Mirror Lake during 1974–84 indicate an annual increase of  $2.9 \mu\text{eq L}^{-1}$  for sodium and  $4.5 \mu\text{eq L}^{-1}$  for chloride. This compares with annual increases of  $2.6 \mu\text{eq L}^{-1}$  for sodium and  $3.0 \mu\text{eq L}^{-1}$  for chloride, based on the 1985–1994 data. The smaller annual increase in chloride for the latter period also likely is influenced by the dilution that occurred following the very wet period during 1990.

## 6. Summary and Conclusions

(1) Road salt moves to Mirror Lake primarily via the northeast inlet stream. However, less than three percent of the 44.6 metric tons of salt applied each year to I93 within the Mirror Lake watershed enters the lake via the northeast stream. A zone of contaminated ground water directly beneath the streambed, at least 0.6 m. thick, also may transfer road salt to the lake, but at a much slower rate. Ground water 5 m on either side of the stream does not appear to be contaminated with road salt, except for an area directly west of the stream extending from the lake to at least 40 m upstream. Road salt evidently is not seeping through a broad reach of the highway diversion berm, but appears to be seeping beneath a portion of the berm, discharging near the headwaters of the northeast inlet stream.

(2) Chloride concentrations in the northeast stream decrease downstream from the diversion berm, indicating that, during winter and spring, uncontaminated ground water discharges into the stream and dilutes the stream water. However, gradients between wells and the stream along two transects perpendicular to the stream are away from the stream during summer months, preventing this dilution during the summer. Consequently, chloride levels commonly reach a maximum concentration in the stream during the late summer months, when gradients reverse and salt-laden water is flushed back into the stream.

(3) Water-table depressions adjacent to the northeast inlet stream caused by transpiration, along with more highly conductive porous media near the lower reach of the stream, have led to contamination of ground water adjacent to the stream on the side of the stream opposite the interstate highway. This contaminated ground water enters the lake along an approximately 70-m-wide shoreline segment that is centered near the stream inlet. Concentrations in the contaminated ground water are as much as an order of magnitude greater than concentrations in uncontaminated ground water.

(4) It is likely that cation exchange takes place between the ponded water east of the berm and the seeps west of the berm, which causes a reduction of sodium and

an increase in calcium, relative to the ponded water, in the stream water directly west of the berm. Little additional cation exchange takes place downstream of the berm.

(5) Chloride concentrations in nearly all wells between I93 and Mirror Lake are large compared to chloride concentrations west of the lake. Steep hydraulic gradients indicate ground water flow toward the lake along most of the eastern shoreline of the lake. Large chloride concentrations are pervasive in the bedrock adjacent to I93, to a depth of at least 123 m, but large chloride concentrations were not pervasive in the glacial material above the bedrock.

(6) Sodium and chloride concentrations were large along a 50-m segment of the easternmost shoreline of Mirror Lake, and along a 70-m segment of shoreline centered at the northeast inlet. Concentrations of sodium and chloride were only slightly larger than normal at the easternmost shoreline segment, but near the northeast stream they were more than 10 times the concentrations of sodium and more than 40 times the concentrations of chloride found along uncontaminated shoreline segments. Slightly-above-normal concentrations of sodium and chloride along the southern portion of the eastern shoreline of Mirror Lake were about equal to concentrations in the lake water. Therefore, it is suspected that occasional reversals in the direction of seepage have contributed to greater levels of sodium and chloride in ground water adjacent to the southern end of the eastern shoreline.

(7) Half of all the chloride that enters the lake comes from the northeast inlet, which contributes only 3% of total streamflow into the lake.

(8) If current rates of increase in concentration of salt were to continue in the northeast inlet and in the lake, the concentration of sodium in the lake would triple and the concentration of chloride would increase by 8 times by the yr 2025, relative to pre-highway concentrations.

### **Acknowledgments**

The authors thank D. Tunnell for collecting lake-shore hydraulic and water-quality data, and J. Crowdes and E. Morency for collection of stream water and wellwater samples. N. Caraco, C. Driscoll, G. Granato, J. LaBaugh and W. Martin provided helpful suggestions for improvement of the manuscript. The authors also thank the USDA Forest Service for allowing access to the Hubbard Brook Experimental Forest. The Hubbard Brook Experimental forest is operated and maintained by the Northeastern Forest Experimental Station, USDA Forest Service, Radnor, PA. Financial support was provided by the National Science Foundation and the A.W. Mellon Foundation.

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