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Major Influences on Lake Water Chemistry in Maine

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ABSTRACT

The chemistry of Maine lakes is highly variable, both on a regional and local scale. The chemistry of lake water depends on both natural and anthropogenic factors. Bedrock lithology is the most important controlling variable. The presence of only small amounts of free carbonate-bearing minerals results in relatively high values for pH, dissolved Ca^{+2}, and HCO_3^-alkalinity. However, proportions of base cations (Ca^{+2}, Mg^{+2}, K^+, Na^+) may vary appreciably even within a terrain dominated by a single mapped lithic unit. Increasing soil thickness or decreasing soil particle size generally results in higher alkalinity, pH, and concentrations of base cations for drainage lakes. Seepage lakes have lower concentrations of base cations and typically are more acidic than drainage lakes. Dissolved organic matter from the decay of vegetation contributes to the acidity of all lakes, but is particularly important in lakes with short water residence times. Marine aerosols significantly affect the chemistry of all lakes within about 50 km of the coast, resulting in elevated Na^+, Mg^{+2}, and Cl^- concentrations. Acidic precipitation and dry deposition of acidic aerosols affects all lakes in differing ways including (1) increasing base cations - especially Ca^{+2} and Mg^{+2}, (2) decreasing alkalinity and pH, (3) increasing Al, and (4) increasing SO_4^{-2}. Lakes are affected by acidic deposition in the order high elevation lakes > lowland seepage lakes > lowland drainage lakes. The impact of non-agricultural watershed activities on water quality are poorly known except for road salting which has increased the concentration of Na^+ and Cl^- appreciably in many lakes.

INTRODUCTION

An estimated 6,000 lakes in Maine are larger than 1 acre (Maine Department of Environmental Protection, 1986). The chemistries of these lakes are determined largely by various geologic, geochemical, geohydrologic, and anthropogenic factors which interact in complex manners. Many of the controls on chemistry can be studied in isolation by stratifying lakes with respect to many of these processes, making significant interpretation possible. This paper focuses on the statistical characterization of Maine lakes in terms of standard limnological parameters and chemistry and then describes several of the major controls on the chemistry of the lakes.

Several studies of lake waters in Maine have been conducted to elucidate pollution problems, facilitate fisheries management, and most recently to understand the effects, real and potential, of acidic precipitation (see e.g. Haines and Akielaszek, 1983). The results are generally not representative of the entire population of lakes in Maine. The studies do, however, enable researchers to focus on and answer specific questions that ordinarily could not be addressed by a broad survey. We do not discuss here the chemical perturbations caused by the activities of man within individual lake basins such as forestry practices and cultural eutrophication. Also, we do not deal with the nutrient status of lakes. Both of these topics have been discussed extensively in other studies (see e.g., Davis et al., 1978; Cowing and Scott, 1975, 1976, 1977). Trace metals in lake waters have also been studied for a variety of reasons (see Norton et al., 1981; Turekian and Kleinkopf, 1958), but we do not deal with them in this paper. Instead, we concentrate on the constituents that typically comprise more than 95% of the dissolved cations and anions in Maine surface waters and focus on processes modifying these parameters. These are: cations: H^+, Ca^{+2}, Mg^{+2}, Na^+, K^+, $Al(OH)_n^{+3-n}$; anions: HCO_3^-, Cl^-, NO_3^-, SO_4^{-2}, and organic anions.

LAKE ORIGIN

Although many lakes in Maine are regulated at their outlet, there are few artificial lakes. The abundance of natural lakes is attributable to the activity of ice and periglacial processes during the Quaternary. Most of Maine's lakes are in bedrock depressions developed by glacial activity. Lakes located in the folded and faulted metasedimentary terrain typically have their long dimension parallel to the regional strike (Davis et al., 1978). Lakes situated in igneous terrain (primarily granites) tend to have their long dimensions oriented parallel to the predominant ice-movement direction, generally northwest-southeast. Soil precursors range from various types of till and stratified material, to marine clays, depending on the location within the state (Thompson and Borns, 1985). About 5 percent of the lakes lie in stratified glacial material, including eskers, kames, outwash, and deltas. These are commonly seepage lakes, i.e. without permanent inlets or outlets.

CONTROLS ON LAKE CHEMISTRY

There are a number of important controls on the major dissolved constituents in surface waters in Maine (Table 1). In most cases these processes are not totally independent from each other, either spatially or mechanistically. For example, the influence of marine salts generally occurs spatially in the same approximate area as does the influence of marine clays (the Presumpscot Formation) on development of soils and hydrology. The contributions of these various factors to the chemistry of Maine lakes are discussed below.

Bedrock Lithology

Most soils in Maine are formed on till and thus generally have mineralogy closely resembling the underlying bedrock. Consequently, bedrock lithology is generally the most reliable guide in predicting the major ion chemistry of surface waters in Maine. In some areas underlain by the Presumpscot Formation or glacial-fluvial deposits, soil materials may be very different in composition from the underlying bedrock. In this section, we consider mineral weathering reactions; in later sections specific soil chemical processes will be discussed.

Unpolluted precipitation is normally mildly acidic (Table 2). Additional CO_2 (as carbonic acid) and organic acids are added to precipitation in the soil profile. Thus the soil solution pH may be depressed below 4.0. Acidic precipitation in Maine adds to the total acidity of the soil solution. These solutions interact with soil and bedrock in two important ways: ion-exchange and chemical weathering. Over long periods of time, chemical weathering is the rate-limiting process which determines the chemistry of water leaving the soil. Short term chemical variations lasting hours to days are more closely related to ion-exchange reactions.

TABLE 1. MAJOR CONTROLS ON LAKE WATER CHEMISTRY IN MAINE, EXCLUDING CULTURAL EUTROPHICATION.

	Geology	Sea Salts	Atmospheric Pollution	Soils	Hydrology	Vegetation	Watershed Activities
pH (H^+)	V		V	V	V	V	V
Ca	V			V	V	V	V
Mg	V	V		V	V		V
Na	V	V		V	V		V
K	V			V	V	V	
Al	V		V	V	V		
HCO_3	V		V	V	V	V	V
Cl		V					V
SO_4		V	V	V	V		
Organic anions				V	V	V	V
F	V			V			

TABLE 2. APPROXIMATE MEAN PRECIPITATION CHEMISTRY FOR MAINE IN 1984 BASED ON NADP DATA (1986) AND ESTIMATED BACKGROUND CHEMISTRY (GALLOWAY ET AL., 1982; BRAKKE ET AL., 1989). EXCEPT FOR PH, VALUES ARE μeq/l.

	pH	H^+	Na^+	K^+	Ca^{+2}	Mg^{+2}	NH_4^+	HCO_3^- Alk.	Cl^-	SO_4^-	NO_3^{-2}	$\frac{Na^+}{Cl^-}$
1984	4.5-4.6	25-30	5-50	<1	2-6	3-12	10	-30	4-58	26-33	12-14	0.86
1774	5-6	1-10	5-50	<1	2-5	3-12	<1	<5	4-58	<1-6	<2	0.86

The reactions of soil minerals with acidic solutions are the chief factors ameliorating the effects of acidic soil solutions. Three equations demonstrate the range of chemical weathering reactions:

(1) $SiO_2 + 2H_2O = H_4SiO_4$
(quartz + water = aqueous silicic acid)
(2) $2KAlSi_3O_8 + 2H_2CO_3 + 9H_2O =$
(orthoclase + acid + water =
$Al_2Si_2O_5(OH)_4 + 2K^+ + 4H_4SiO_4 + 2HCO_3^-$
kaolinite + aqueous K + aqueous silicic acid + bicarbonate)
(3) $(Ca,Mg)CO_3 + H^+ = (Ca^{+2},Mg^{+2}) + HCO_3^-$
(calcite + acid = aqueous ions + bicarbonate)

Reaction (1) neither produces nor consumes acidity during chemical weathering. Reactions similar to (2), typical of aluminosilicate mineral weathering, consume acid and produce bicarbonate alkalinity (HCO_3^-), release base cations (Ca^{+2}, Mg^{+2}, K^+, and Na^+) to solution, and produce clay. The extent of this reaction depends largely on the time of reaction in the soil. The equilibrium pH for many such silicate reactions is over 8. Generally, however, the residence times for water in soils are less than is required for equilibrium to be achieved. The third reaction also has an equilibrium pH greater than 8, but the kinetics are rapid in contrast to reaction (2). Consequently, terrain with bedrock or soils containing free carbonate minerals such as calcite or dolomite, as in some of the stratigraphic members of the Seboomook Formation in northern Maine (see below), may have waters with pH over 8 and high concentrations of Ca^{+2} and Mg^{+2}. Thus, due to the kinetics of these weathering reactions, igneous rock terrain typically yields waters influent to lakes with lower pH and lower concentrations of base cations than does a terrain underlain by heterogeneous sedimentary and metasedimentary rocks.

Precipitation Chemistry

Acidic Deposition. The composition of wet precipitation and dry deposition is also a major influence on surface water chemistry. The approximate composition of wet deposition in Maine is being well characterized by a system of collectors located at Greenville (Fernandez et al., 1986), North Bridgton, Mount Desert Island, Presque Isle, and Caribou as part of the national network (NADP, 1986). Typical precipitation chemistry is given in Table 2; H^+, SO_4^{-2}, and NO_3^- are the dominant ions.

Sulfate is the dominant anion in many surface waters in Maine. The most important source for SO_4^{-2} is atmospheric deposition. Sulfate derived from oxidation of sulfide minerals in the bedrock or soils is relatively unimportant by comparison except in a few scattered locales. Because of the acidity of modern precipitation, the pH and alkalinity of lake waters are also partly determined by wet and dry deposition of acidic or acidifying substances from the atmosphere.

The measured wet component of atmospheric deposition does not include deposited gaseous components, dry fall of particulates, or wet deposition of material which would not produce measurable precipitation in a collector. There are no regional measurements of dry deposition in Maine, and thus their contribution can only be estimated. Dry deposition of gases (principally SO_2 and NO_x) and passive interception of fog and cloud moisture may contribute from nearly zero to as much as 50 percent of the total acid anions delivered to Maine lakes (Norton et al., 1988a). This level of input is suggested by a comparison of atmospheric deposition rates of conservative elements (e.g. Cl, and to a lesser extent S) and their concentrations in lakes and streams. Areas with rough topography and non-deciduous forest stands receive the most dry deposition. Lake surfaces directly receive the least.

Prior to pollution of the atmosphere, the major difference between modern and pre-pollution precipitation chemistry would have been the comparatively low concentrations of SO_4^{-2}, NO_3^-, NH_4^+, and H^+ (Table 2). Maine soils and ecosystems are presently N-limited. This is indicated by the observation that little N is transported to ground water or surface water except during periods of high flow. The NO_3^- is metabolized and stored in organic matter and the H^+ is neutralized by the reduction of NO_3^-. If Maine systems were to become saturated with NO_3^-, as they appear to be in some forested areas of eastern and western Europe and southern Norway (see e.g. Henriksen, 1987), NO_3^- in precipitation would then have an acidifying influence similar to that of SO_4^{-2}.

Sea Salts. Aerosols derived from the ocean are an important constituent of wet precipitation and comprise a large fraction of the dry deposition near the coast. The Na^+:Cl^- equivalent ratio in precipitation near the coast is close to 0.86 (NADP, 1986), the value in sea water. On this basis, it is assumed that sea salts in precipitation are present in essentially the same proportion as they are in sea water. If there are no other sources of Cl^- to surface waters, it is possible to "correct" a lake water chemical analysis for those constituents derived from sea salt deposition. The sea salt contribution to the other constituents is subtracted in their proper proportions to Cl^- in seawater. The residual values are termed "excess" or "non-marine" (designated * in the text and tables) and with the exception of sulfate, are largely derived from chemical weathering in the drainage basin.

A consequence of the deposition of marine salts either in dry or wet deposition is the existence of a "sea salt effect" (Wiklander, 1975) in soils (Kahl et al., 1985; Norton et al., 1987b; Wright et al., 1988). This process involves the ion exchange of Na^+ and Mg^{+2} for Ca^{+2} and H^+ in soils with a subsequent lowering of alkalinity and pH in the runoff (Equation 4). If pH declines below 5, Al may be exchanged from the soil or mobilized from stream substrates as well (Norton et al., 1987a). This phenomenon may episodically influence the pH of small lakes and streams, and certainly the Na:Cl ratio in streams and lakes near the ocean.

Soils

Most runoff water is routed through near-surface soils. Therefore, reactions in soil normally have the greatest direct influence on the chemistry of surface waters. Consequently, the chemical reactivity of soil (especially cation exchange properties) and hydrologic characteristics of flow through watersheds are controlling factors in determining the temporal chemistry of lake water. Short term variations in soil water chemistry are controlled by cation exchange reactions, principally involving adsorption and desorption of the four major base cations, aluminum, and hydrogen cations coupled with an anion. Reaction 4 schematically depicts the ion exchange of H^+ for the cations adsorbed on a soil particle, with the exchanging cations listed in order of decreasing abundance for a typical acidic soil.

4) $H^+ + [Al^{+3}, H^+, Ca^{+2}, Mg^{+2}, K^+, Na^+]$ -Organic particle
$= [Al^{+3}, H^+, Ca^{+2}, Mg^{+2}, K^+, Na^+] + H^+$ -Organic particle

Coarse, well-drained soils typically yield lower alkalinity waters and a chemistry which is temporally stable. Poorly drained soils, commonly saturated, yield water with greater chemical variation. At low flow, pH and the concentration of dissolved solids tend to be higher. At high flow, water may bypass the soil column entirely, resulting in lower pH and low concentrations of dissolved solids due to dilution. The short term variations in the water chemistry of streams due to these soil and hydrologic factors may significantly alter the chemistry of lakes with short retention times.

Hydrology

Hydrologically, Maine lakes range from the extreme case of perched lakes where the lake acts as a precipitation collector and recharges ground water, to drainage lakes where most of the water leaving the lake by surface flow entered as streams, ground water, or direct surface runoff. For perched lakes, interactions between soils and precipitation are minimal, and the lake chemistry is dominated by precipitation. Most lakes in Maine are drainage lakes. Lakes with long water residence times are less subject to short term variations in water quality. Shallower lakes with large watersheds can have significant episodic variations in chemistry associated with runoff from snow melt and rainstorms.

Vegetation

In undisturbed watersheds, vegetation exerts four major influences on the chemistry of lakes. First, seasonal variations in the uptake and release of limiting nutrients, particularly Ca and K, are reflected in oscillations of the concentration of Ca^{+2} and K^+ in streams and small lakes (Likens et al., 1977). Uptake of NO_3^- and NH_4^+ by terrestrial vegetation and phytoplankton in lakes typically maintains dissolved N at very low concentrations during the growing season. Second, the canopy of a forest acts as a filter, capturing dry deposited gases such as SO_x and NO_x, and particulates such as marine aerosols, thereby increasing the load of constituents to a watershed (Harriman and Morrison, 1982). This effect has been shown to be important in Maine (Norton et al., 1988a). Third, terrain with bog vegetation may yield surface waters which have been depleted of cations by ion exchange of base cations for hydrogen ions. Sphagnum is well known for the ability to acidify water through ion-exchange (Clymo, 1984). Lastly, the release of dissolved organic carbon, either from living biomass or from the decay of dead plant matter, results in the formation of compounds with carboxylic groups. These dissociate to form acid anions, thus partly controlling the acid-base status of lake waters (Cronan and Aiken, 1985). This process also adds color to the water and provides organic ligands for the complexation of metals.

Watershed Activities

Several activities are sufficiently common in Maine to affect the chemistry of numerous lakes. They include agriculture, cultural development, forestry practices, and road salting. The first two activities have been studied and are reported on by Cowing and Scott (1975, 1976, 1977) and Davis et al. (1978). We do not discuss these processes here because we focus on lakes largely unaffected by these processes.

There are no published studies on the effects of forest cutting on stream chemistry in Maine; however, land scarification is known to cause an increase in downstream water temperature, increased run-off, short term elevated export of detritus of various types, and a general increase in the concentration of many dissolved constituents (Likens et al., 1977). The loss of the forest canopy also decreases the capacity of the trees to capture dry deposition, decreasing the input budget for nitrogen, sulfur, and marine aerosols (Harriman and Morrison, 1982).

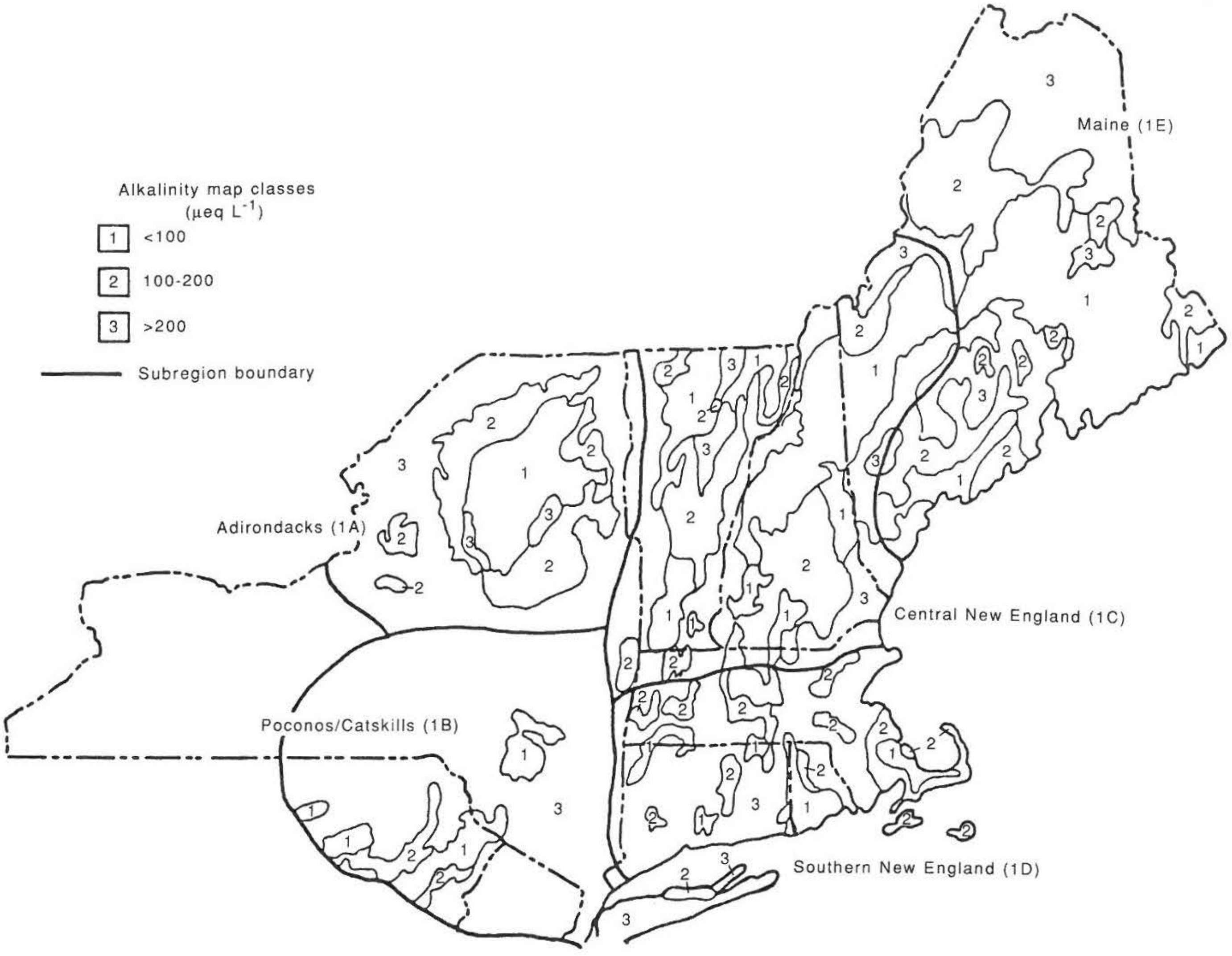

Figure 1. Sub-regions and alkalinity map classes for the Eastern Lake Survey, northeastern region (from Linthurst et al., 1986).

Road salting results in a significant addition of NaCl to lakes and streams downstream from roadways in Maine. Chemical outliers are evident in synoptic data sets because of the general relationship between inland distance and Cl^- concentration of lake waters (Mairs, 1967). Consequently, when an inland lake has abnormally high Cl^-, it can be identified as polluted by road salt, and removed from consideration in the data set. However, near the coast where natural salt loading is higher, the chemical signal from road salting may not be identified as such and consequently the resulting sea salt corrections to the chemistry of a lake may yield inaccurate results. Although there are no data that we know of to demonstrate impacts in Maine, excess salt in stream or lake water which then passes through wetlands could generate a salt effect and acidify the water.

MAINE LAKE CHEMISTRY

EPA Eastern Lake Survey

Considerable concern has been expressed about the effects of acidic precipitation on lakes in the northeastern part of the United States. The U.S. Environmental Protection Agency has been charged with the assessment of the extent of damage to lakes and the resource at risk. The Eastern Lake Survey (ELS; Linthurst et al., 1986) was designed to determine the number and distribution of acidic and low alkalinity lakes in the eastern U.S. The survey design split Maine into two subregions (1E and 1C on Fig. 1). As part of the ELS, 225 lakes in Maine out of an estimated target population of 1,966 lakes with an area greater

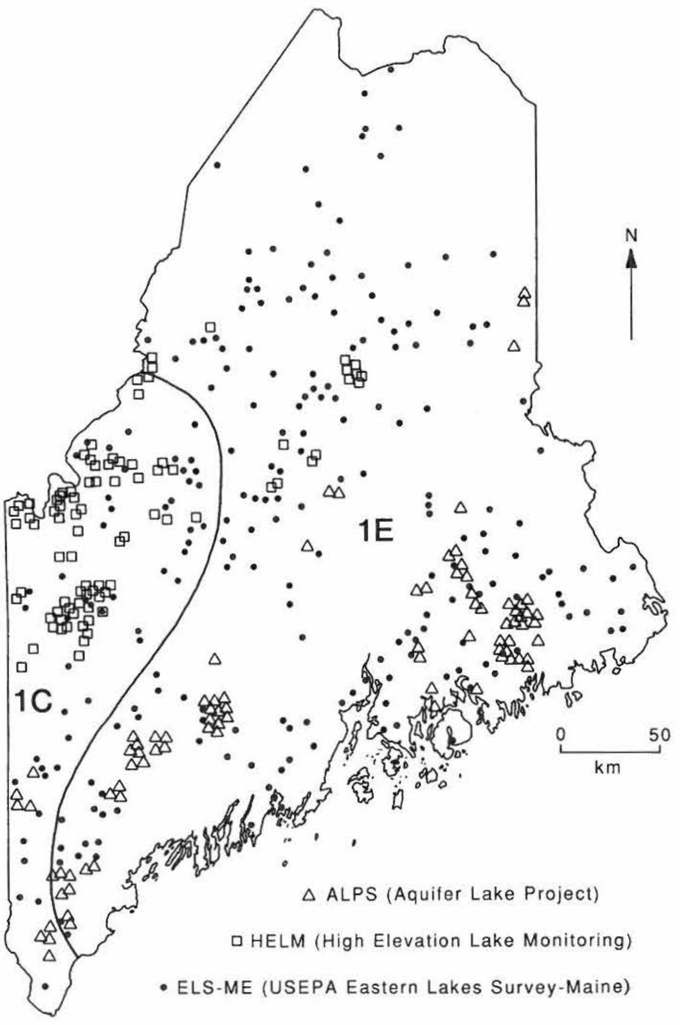

Figure 2. Distribution of lakes sampled in Maine (Regions 1C and 1E) for the Eastern Lake Survey, the ALPS (aquifer lakes) survey, and the HELM (high elevation lakes) survey.

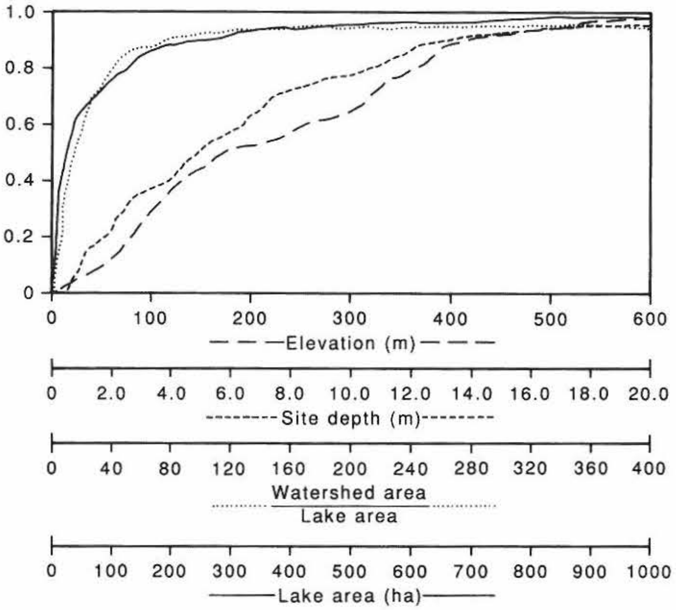

Figure 3. Cumulative frequency distribution of elevation (m), site depth (at the point of water sampling)(m), ratio of the watershed area to lake area, and lake area (ha) for Eastern Lake Survey lakes in Maine.

TABLE 3. PHYSICAL CHARACTERISTICS OF ELS LAKES IN MAINE. 20TH PERCENTILE (Q_1), MEDIAN, AND 80TH PERCENTILE (Q_4) ARE GIVEN.

Parameter	Q_1	M	Q_4
Lake Area (ha)	8.2	23.7	124.1
Depth at sampling site (m)	1.8	4.9	10.6
Elevation (m)	81	170	363
Watershed area (ha)	111	428	2594
Watershed area/Lake area	6.8	15.1	39.9
Flushing Time (y)	0.05	0.23	0.75

than 4 ha were sampled during the fall of 1984 (Fig. 2). Lakes were selected as a systematic random sample from the population of all lakes identified on 1:250,000 scale U.S. Geological Survey maps. The formulation of sub-populations, and methods of sampling and analysis are found in Linthurst et al. (1986). A description of results for the northeast U.S. region (Fig. 1) is given in Brakke et al. (1988). Our analysis of the data has developed population estimates based on all Maine lakes surveyed. Smaller lakes were not included because many are not shown on the small scale maps and it was not possible to develop a uniform selection strategy for small lakes in different areas. These small lakes are an important resource in some areas. They are discussed separately in this paper.

Physical Characteristics of the ELS Lakes. The physical characteristics of the lakes sampled are summarized in Figure 3 and Table 3. Many of the lakes sampled were small, with 20% less than 8.2 ha. Most were lowland lakes; only 11.5% were above 400 m in elevation. A total of 41 lakes (2.1%) were estimated to occur above 600 m. This estimate compares well to the 42 lakes at least 4 ha in area and higher in elevation than 600 m identified by the High Elevation Lake Monitoring (HELM) project (Kahl and Scott, 1987). Most of the ELS lakes had watershed area:lake area ratios less than 20, with 12.9% having ratios less than 5. Over 90% of the lakes are drainage lakes, having an outlet. Assuming that precipitation averages 1 m in the north to 1.4 m along the coast and a 40% evapotranspiration rate, and knowing that the average watershed area:lake area is about 15 and the mean depth is 5 m, the residence time of water in most lakes in Maine is on the order of months to a few years.

Chemical ELS Data. Cumulative frequency diagrams of chemical components for the lakes sampled in Maine by ELS are presented in Figures 4 to 8. Table 4 gives the 20th percentile (Q_1), median, and 80th percentile values for selected chemical parameters in the target population of lakes.

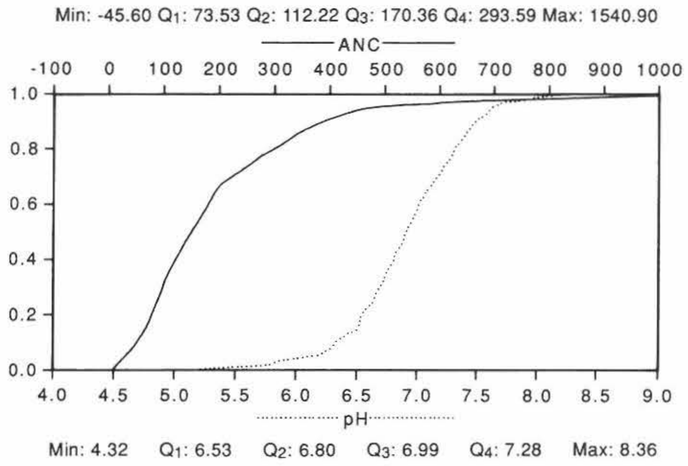

Figure 4. Cumulative frequency distribution of pH and acid neutralizing capacity (ANC) for Eastern Lake Survey lakes in Maine. ANC in µeq/l.

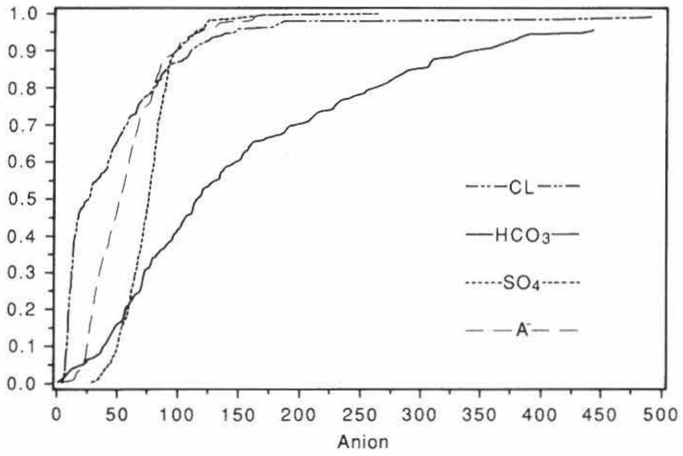

Figure 5. Cumulative frequency distribution of SO_4, Cl, organic anions, and HCO_3 anions for Eastern Lake Survey lakes in Maine. Data in µeq/l.

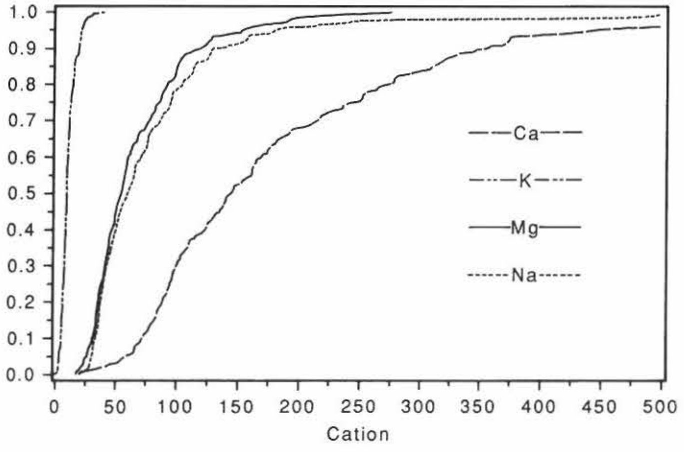

Figure 6. Cumulative frequency distribution of Ca, Mg, Na, and K for Eastern Lake Survey lakes lakes in Maine. Data in µeq/l.

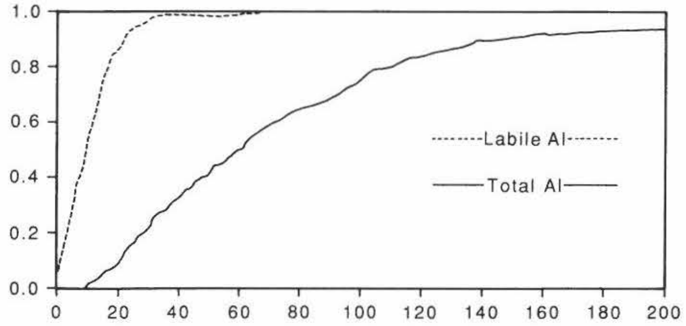

Figure 7. Cumulative frequency distribution of labile Al and total Al for Eastern Lake Survey lakes in Maine. Data in µg/l.

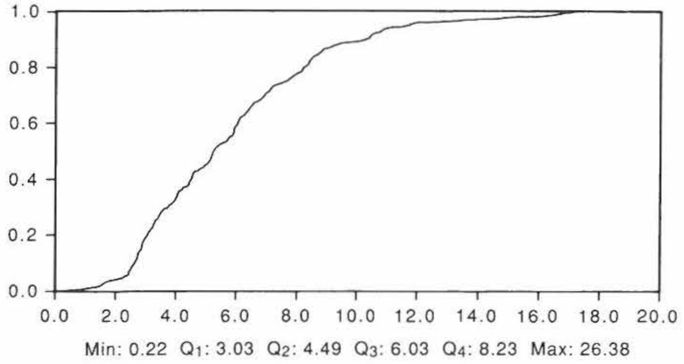

Figure 8. Cumulative frequency distribution of dissolved organic carbon (DOC) for Eastern Lake Survey lakes in Maine. Data in mg/l.

TABLE 4. FIRST QUINTILE, MEDIAN, AND FOURTH QUINTILE VALUES FOR CHEMICAL PARAMETERS FOR ELS LAKES IN MAINE.

		Q_1	Med	Q_4	to Convert µeq/l to mg/l, multiply by
pH		6.53	6.89	7.28	
Ca	µeq/l	88.5	142.6	270.8	0.020
Mg	"	35.4	53.8	91.3	0.012
Na	"	37.5	59.4	103.3	0.023
K	"	6.4	8.9	12.8	0.039
ΣAl	µg/l	28.0	59.4	106.3	
Ionic Al	"	3.2	9.3	17.4	
Alk[1]	µeq/l	69	129	235	0.061
SO4-2	"	59.7	76.5	90.1	0.048
Cl-	"	10.2	26.0	79.3	0.035
F-	"	1.2	1.6	3.1	0.019
NO3-	"	0.0	0.2	0.8	0.062
NH4+	"	0.8	1.3	2.0	0.018
SiO2	mg/l	0.9	2.0	3.4	
DOC	"	3.0	5.2	8.2	

[1] Alk = $HCO_3^- - H^+$

There are environmental gradients in Maine caused by bedrock lithology, deposition of marine aerosols, soil types, climate, and sulfate deposition. Consequently, one can not derive an "average" lake composition for Maine by assuming median values for all the chemical parameters. However, the figures are useful for evaluating the range of values present in Maine lakes and the distribution of lakes within these ranges. As a group, Maine lakes are relatively low in dissolved solids, and have low alkalinity, pH, and specific conductance. Calcium is typically the dominant cation, while the dominant anion may be Cl^-, HCO_3^-, or SO_4^{-2}, depending on the original alkalinity, location within the state along the gradient of marine aerosols, and total anthropogenic sulfate deposition. The resultant chemistry of the population is dominated by chemical weathering, atmospheric deposition, production of organic acidity, and deposition of sea salts.

pH. The long term average pH of a lake is determined by the balance between the release of strong base cations from chemical weathering of minerals and the concentrations of SO_4^{-2}, Cl^-, NO_3^-, HCO_3^-, and organic anions. Because of the large capacity of soil to exchange cations, changes in precipitation chemistry (particularly pH) may not be reflected in proportional changes in runoff chemistry.

Currently, SO_4^{-2} comes largely from atmospheric deposition. In Maine, where present adsorption of SO_4^{-2} by soils is minimal (Rochelle and Church, 1987), SO_4^{-2} is a mobile anion, passing through the soil with little or no reaction with the soil material (Reuss and Johnson, 1985). As SO_4^{-2} passes through the soil, it must be accompanied by a cation. If increases in chemical weathering or ion exchange processes can not fully compensate for the increased flux of SO_4^{-2} through the soil, the system will acidify, possibly mobilizing Al from the soils if the pH is less than about 5.

Chloride ions from marine aerosols are accompanied largely by the cations Na^+ and Mg^{+2}. In episodic salty precipitation events, Na^+ and Mg^{+2} may exchange for Ca^{+2}, H^+, and Al^{+3} (if pH is sufficiently low), producing the strong acid HCl. Because nitrogen is a limiting nutrient in Maine ecosystems, most of the NH_4^+ and NO_3^- is consumed biologically. However, during periods of high precipitation or during snow melt, NO_3^- may not be assimilated by the system as rapidly as it is delivered; if not, then it passes through the system as an acidic anion. Both Cl^- and NO_3^- generally play a small role in determining the present pH status of Maine lakes, but they may be important in influencing the pH of individual lakes or streams over short periods (Kahl et al., 1985; Wright et al., 1988).

Bicarbonate (HCO_3^-) is present in surface waters as a result of the dissociation of H_2CO_3 and/or the weathering of carbonate minerals. The former is a product of H_2O and CO_2 equilibration with atmospheric and elevated ground water CO_2 pressure, the latter caused by respiration in the soil. As acid is consumed by chemical reactions (equations 2, 3, and 4), pH rises and more H_2CO_3 dissociates to yield HCO_3^-. In regions without acidic precipitation, clear water lakes with dissolved organic carbon less than about 2 mg/l, no sea salts, and no SO_4^{-2} from polluted precipitation, HCO_3^- is the only anion of significance. In this case, the concentration of HCO_3^- represents the amount of chemical weathering that occurred in the watershed.

The ELS in Maine contained the smallest percentage of acidic lakes (pH less than 5) of all states in the northeastern United States. Only 0.4% and 4.6% of the target population were inferred to have a pH less than 5 and 6, respectively (Fig. 4), although data from other University of Maine and Maine Department of Environmental Protection studies indicate that between 1 and 2% are acidic when all size classes are included.

Alkalinity and Acid-Neutralizing Capacity. A definition of alkalinity is:

(5a) $\text{alkalinity} = (HCO_3^-) - (H^+ + 3\text{-}nAl(OH)_n^{3-n} + \text{other less abundant weak acid cations})$

where all concentrations are in moles.
Above pH = 5, charged Al is nearly zero and the relationship can be simplified to:

(5b) $\text{alkalinity} = (HCO_3^- - H^+)$

Below pH = 5, HCO_3^- is essentially zero and

(5c) $\text{alkalinity} = -(H^+ + 3Al^{+3} + \ldots)$

In a system with only HCO_3^- as an anion, and prior to any chemical weathering or ion exchange, alkalinity equals zero and is independent of CO_2 pressure in surface waters. The release of cations from the soil produces positive alkalinity in amounts equal to the cations released. If the alkalinity is less than the base cations released, other anions must be present for ion balance. If these other anions are weak acid anions, they will contribute to the total acid neutralizing capacity (ANC) of the solution. ANC must, therefore, be greater than or equal to alkalinity. Strong acids, fully dissociated at normal pH, decrease alkalinity and may even produce negative values.

Weak acids are associated with dissolved organic matter, expressed as dissolved organic carbon (DOC). Organic anions, formed by the dissociation of dissolved organic material, contribute to the acidity status of natural waters in Maine. If the production of H^+ from the formation of organic anions is not accompanied by an equivalent export of cations, the water is naturally acidic.

Figure 4 depicts the ANC relationships for Maine lakes. There were 10.2% of the lakes with alkalinity below 50 µeq/l; 67% were below 200 µeq/l. Figures 4 and 5 and Tables 5 and 6 show that ANC is less than the total anionic charge for Maine lakes, with Cl^-, SO_4^{-2}, and organic anions responsible for the remainder.

Major Cations. Most of the cations in Maine lake water (Fig. 6) are derived from weathering of the bedrock. The exception is Na^+ and to a lesser extent Mg^{+2} and Ca^{+2} in coastal lakes. Minor amounts in all lakes are derived from sea spray and soil dust in precipitation. The contribution of marine aerosols to the cation content of lakes can be estimated from the concentration of Cl^-, assuming that cations are contributed in the same proportion as they are present in sea water. The corrected values for cations and anions (designated *) are called sea salt corrected (non-marine) and represent the concentrations derived from the watershed and SO_4^{-2} derived from the watershed or atmosphere. The order of concentration of the cations in lakes is typically $Ca^{+2} > Mg^{+2} > Na^+ > K^+$. For lakes in temperate regions in the northern hemisphere, Ca^{+2} and Mg^{+2} derived from chemical weathering typically comprise 70-90% of the total base cations (corrected for sea salts) in geologic terrain with no free carbonate minerals (Wright, 1983). Consequently, there is a strong relationship between the sum of $(Ca^{+2} + Mg^{+2})$ and alkalinity (compare Figs. 5 and 6, Equation 3), except where organic acids and/or SO_4^{-2} play a major role in the ion balance.

Ca and K are biologically important elements. Biological uptake of these elements in the drainage basin during the growing season, and release during leaf senescence may impart a seasonal oscillation to their concentrations. This is especially true in lakes with short flushing times.

There are three major sources of Na^+ for Maine lakes: (1) weathering of geologic materials, which typically yields Na^+ concentrations that are lower than those of Ca^{+2} or Mg^{+2} (Fig. 6, Tables 5 and 6); (2) road salting, which adds primarily NaCl to the water (in extreme examples, Na^+ from road salting exceeds the contribution from chemical weathering); and (3) marine aerosol inputs, which may dominate the chemistry of coastal lakes. Sodium may be the dominant cation in lakes within 10-15 km of the ocean (e.g. lakes 1 and 2 in Table 5).

Aluminum. Dissolved Al has been implicated as one of the critical parameters controlling chronic and acute toxic effects to fish in acidic waters (Baker and Schofield, 1982). Al may be complexed with organic (DOC) or inorganic ligands (such as F^-, SO_4^{-2}, and OH^-) or it may be present as Al^{+3}, depending on pH and the availability of complexing agents. The $Al(OH)_2^+$ and $Al(OH)^{+2}$ species are believed to be the forms most toxic to fish.

Virtually all the dissolved Al in lake waters is derived from geologic sources. Although Al is abundant in most rocks and soils, dissolved Al is present in relatively low concentrations because of the low solubility of Al-bearing phases in soil. Below pH 5, however, the solubility of Al phases increases. Figure 7 depicts the total Al concentration (all forms) and the concentration of charged Al species (extractable Al). Effects on fish may occur with extractable Al values as low as 50 µg/l. For the ELS Maine data, only 1% of the lake population had values of 50 µg/l or more at the time of sampling.

Anions. Virtually all the Cl^- in lake waters in Maine is derived from sea salts in precipitation, except where road salt drainage enters lakes. Consequently, there is a strong relationship between Cl^- concentration and distance to the coast (Norton et al., 1988a). The geochemical inertness of Cl^- makes it suitable as a tracer for marine aerosol inputs. The concentration of Cl^- allows for estimation of SO_4^{-2} inputs in precipitation from marine versus non-marine sources.

With the exception of a few polluted systems, Maine lakes have SO_4^{-2} concentrations between 40 and 110 µeq/l (Fig. 5). There are several possible reasons for local variation. First, wet SO_4^{-2} deposition varies seasonally (NADP reports). Consequently, lakes with short water residence times will reflect more recent precipitation events. Second, some lakes may have sulfate reduction taking place either within the hypolimnion or at the sediment-water interface, although this effect appears to be minimal on a regional basis (Norton et al., 1988b). Third, the amount of dry deposition in contiguous drainages can differ significantly because of lake hydrologic type, topographic relief, the aspect of the land surface, and the type of vegetative cover. Seepage lakes, which may in the extreme case have a watershed only as large as the lake itself (a perched seepage lake), receive very small contributions of dry deposition and typically have a SO_4^{-2} concentration 20 µeq/l less than neighboring lakes. Lakes 3 to 7 of Table 5 are all located within 4 km of each other and yet have SO_4^{*-2} concentrations ranging from 46 to 88 µeq/l. The low values are associated with small drainage basins which have little topographic relief.

The fourth possible cause of local variation in SO_4^{-2} is sources of geologic sulfur within a drainage basin. A number of lithologic units in Maine (e.g. the Penobscot Formation) contain various sulfide minerals. When sulfide-bearing rocks are freshly exposed, oxidation of the sulfide minerals occurs, producing H_2SO_4. Reactions are rapid, although typically the weathering rind formed causes a dimunition in the oxidation rate with time. Postglacial soils in Maine are sufficiently old (11,000 to 13,000 years) that unoxidized sulfide minerals are relatively rare in well drained soils. The contribution of geologic SO_4^{-2} to lake waters is probably not easily detected, given the magnitude of the atmospheric flux. In the entire ELS Maine data set, only the lake (First Pond) downstream from the Black Hawk mine at Blue Hill appears to be affected by geologic SO_4^{-2}.

Regional and local variation in SO_4^{-2} in Maine lakes has been assessed by Norton et al. (1988a). Values for SO_4^{-2} in lakes in the western half of the state averaged about 10 µeq/l higher than the state median of 77 µeq/l, whereas lakes east of longitude 69 and south of latitude 46 had SO_4^{-2} values about 10 µeq/l below the state average. Lakes very close to the coast have elevated SO_4^{-2} associated with sea salts. There is only minor spatial variation in wet deposition of SO_4^{-2}. In 1984, the SO_4^{*-2} concentration in wet deposition ranged from 33 µeq/l at Bridgton to 26 at Acadia National Park (NADP, 1986). This flux is supplemented by locally and regionally variable dry deposition. The total atmospheric flux is then concentrated by evapotranspiration to yield lake SO_4^{-2} values which average nearly three times the wet precipitation values (Fig. 5). Variation in the regional averages is believed to relate primarily to variation in dry and occult deposition (Norton et al., 1988a).

The extra SO_4^{*-2} in lake water is related to higher SO_4^{-2} concentrations of modern precipitation over pre-industrial values

(Table 2). This increase implies that there is an excess of acid going through the system. This extra flux of anions through lakes may have been associated with elevated chemical weathering or ion exchange to release base cations, or an increased flux of H^+ to the lake.

Dissolved Organic Carbon (DOC). Figure 8 shows the distribution of DOC in the sampled ELS lakes in Maine. DOC is related to water color by the empirical relationship for region 1E:

6) DOC = 0.096(Pt-Co units) + 2.46 (Linthurst et al., 1986)

Thus lakes with higher DOC typically have perceptible brown color. There is also a general relationship between increased DOC and increased concentrations of organic anions, as calculated by ion balance difference. DOC concentrations are determined by the rate of production of DOC, dilution by rainfall, residence time of water in lakes (Brakke et al. 1987a,b), and perhaps other factors. Thus there are regional and seasonal variations. For surface waters, each mg of DOC contributes about 5 μeq/l of dissolved organic anions to the solution. The contribution of DOC to anion concentrations and to color for lakes where DOC is less than 3 mg/l appears to be somewhat less than predicted by Equation 6, and by the empirical relationship of 5 μeq/l/mg DOC.

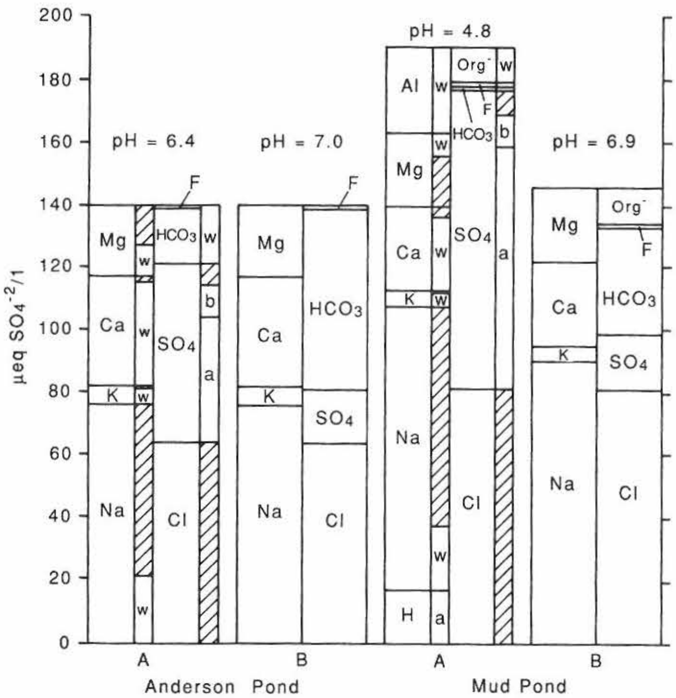

Figure 9. Ion balances for Anderson and Mud Ponds, Maine with (A) and without (B) excess SO_4^{-2}. Ions labeled w are derived from the watershed; ions labeled a are anthropogenic (excess) SO_4^{-2}. Background SO_4^{-2} is assumed to be 10 μeq/l. Shaded area in A = marine aerosol contribution.

CASE STUDIES

We have selected a number of example lakes from the ELS data set to illustrate how the major processes discussed above influence lake chemistry. The lakes have been chosen on the basis of having few independent variables operating on them so as to isolate the effects of the specific processes. The chemistry of the lakes is not static and thus interpretation of a single data point in time must be done with some caution. Variations in meteorology, seasonality of processes, and intervention by man can cause variation in the chemistry of a lake, but most of the information on basic major ion chemistry can be gained from a single, well characterized sample.

Acidity in Maine Lakes

The term "acidic" refers to a current state of a lake and indicates nothing about how a lake became acidic. In contrast, "acidification" is a process resulting in a change of state. From the perspective of acidic precipitation, the concern is with rapid acidification (less than a 100 year time-frame) related to the input of increased amounts of strong acid from the atmosphere due to acidic precipitation. Acidification may be manifested in a number of ways, including a decrease in alkalinity, lowering of pH, increased desorption of cations and chemical weathering, mobilization of elements such as Al from terrestrial and aquatic parts of the ecosystem, and decreased DOC. Therefore, acidification can, but does not necessarily, produce acidic lakes (pH less than 5 or ANC less than 0).

We present an interpretation of two lakes, one acidic and one not, both of which appear to have lost some ANC (been acidified) (Fig. 9). Anderson and Mud Ponds are both located on the Tunk Mountain pluton near Cherryfield. The marine aerosol corrected values for the base cations (Table 5) are typical of many lakes situated on granite.

The effect of the input of H_2SO_4 in precipitation and dry fall can be assessed by considering the chemistry of the two lakes. The ion balances for the two ponds are shown in Figure 9 (column A). Prior to acidic precipitation (Table 2), $SO_4{*}^{-2}$ is assumed to have been largely absent from the lakes. Background pre-pollution SO_4^{-2} has been estimated at about 10 μeq/l by Brakke et al. (1989). Removing the excess SO_4^{-2} (i.e. $SO_4{*}^{-2}$from the analysis necessitates that the charge balance be maintained by some combination of removal of cations or an increase in anions. There is no basis for substantially changing the organic acid content or Cl^- (from sea salt) content of the water over time. The simplest resolution of the charge problem is to replace the $SO_4{*}^{-2}$ with HCO_3^- (alkalinity)(column B) on an equivalent basis. The calculated alkalinity (58 μeq/l) then nearly equals the sea salt corrected base cations (69 μeq/l) and the pre-pollution pH of Anderson Pond can be calculated as about 7.0. This pH is a maximum estimated value and suggests the maximum amount of acidification that could have occurred. Using the methods of

TABLE 5. WATER CHEMISTRY OF SELECTED LAKES IN MAINE, LOCATED ON GRANITE.

	Lake ID	Lake Name	pH	H^+ µeq/l	Na	Na*	K	K*	Ca	Ca*	Mg	Mg*
1	IEI-101	Long Pond-Isle Au Haut	6.76	0.2	498	23	15.2	4	82.7	61	127	16
2	IEI-079	Upper Hadlock Pond	6.93	0.1	211	57	7.2	6	92.8	85	40.7	5
3	IEI-131	Anderson Pond	6.38	0.4	76.3	21	5.8	5	35.2	33	22.8	10
4	IEI-132	Little Long Pond	6.08	0.8	91.1	21	7.2	6	40.2	37	27.1	11
5	IEI-133	Tilden Pond	6.91	0.1	96.4	34	7.4	6	59.2	56	30.4	15
6	IEI-134	Mud Pond	4.76	17.4	90.6	21	4.8	3	26.9	24	23.5	7
7	IEI-135	Salmon Pond	7.00	0.1	90.0	28	7.4	6	66.4	63	31.4	16
8	IEI-136	Spring River Lake	6.55	0.3	127	26	7.6	5	51.2	46	32.9	10
9	IEI-126	Fitts Pond	6.75	0.2	69	23	8.6	7	79.6	78	27.9	19
10	IEI-012	Hopkins Pond	6.95	0.1	62.1	27	7.8	7	83.6	82	31.8	23
11	IEI-058	Lower Sebago Lake	7.14	0.1	80.6	44	16.9	16	88.5	86	32.5	24
12	IEI-020	Second Debsconeag Lake	7.15	0.1	52.4	40	13.5	13	138	137	32.7	29
13	IEI-030	Lower Jo-Mary Lake	7.01	0.1	40.2	29	9.5	9	91.5	91	28.4	26

	Lake ID	Lake Name	Cl	SO_4	SO_4*	NO_3	HCO_3	DOC mg/l	ANC µeq/l	F	ΣCations* µeq/l	$\frac{\Sigma Cat^*+Mg^*}{\Sigma Cations^*}$
1	IEI-101	Long Pond-Isle Au Haut	552	143	88	0.1	39.5	2.42	34	3.7	104	0.74
2	IEI-079	Upper Hadlock Pond	179	95.5	77	0.4	56.2	3.07	51	3.9	153	0.59
3	IEI-131	Anderson Pond	64	57.3	51	0.9	17.3	1.98	10	1.3	69	0.62
4	IEI-132	Little Long Pond	81.6	78.1	70	0.1	8.5	1.39	6	1.9	75	0.64
5	IEI-133	Tilden Pond	72.7	57.2	50	0.0	50.9	2.60	47	4.2	111	0.64
6	IEI-134	Mud Pond	81.4	96.1	88	0.0	0.9	2.82	-19	1.5	55	0.56
7	IEI-135	Salmon Pond	72.6	53.5	46	0.0	57.0	2.82	56	4.1	113	0.70
8	IEI-136	Spring River Lake	117	73.9	62	0.0	28.5	1.69	20	3.5	87	0.65
9	IEI-126	Fitts Pond	46.4	92.6	88	0.5	36.9	1.77	30	21.2	127	0.76
10	IEI-012	Hopkins Pond	41.2	71.1	67	0.0	58.4	2.83	52	9.0	139	0.76
11	IEI-058	Lower Sebago Lake	41.6	48.8	45	0.3	82.7	6.05	86	4.6	170	0.65
12	IEI-020	Second Debsconeag Lake	14.1	83.0	82	0.3	113.2	2.27	118	20.9	219	0.76
13	IEI-030	Lower Jo-Mary Lake	12.9	58.9	57	0.0	71.4	3.54	73	4.5	155	0.75

Sea salt corrections: Na*= Na - 0.86 Cl; K* = K - 0.02 Cl; Ca* = Ca - 0.04 Cl; Mg* = Mg - 0.2 Cl; SO_4* = SO_4 - 0.1 Cl

Wright (1983), a lower alkalinity and pH of about 6.8 are calculated.

Today Mud Pond (Fig. 9) has more SO_4^{-2}, negative alkalinity, lower pH, and higher dissolved Al than Anderson Pond. Following the Anderson Pond example, removal of the excess SO_4^{-2} results in restoration of positive alkalinity, loss of all ionic aluminum and a calculated pre-pollution pH of about 6.9. Mud Pond has become acidified because it receives about 37 µeq SO_4^{-2}/l more than Anderson and the rate of alkalinity production (as expressed by the watershed-originated base cations) is 13 µeq/l less. This leads to a net difference in acidity of the two systems of about 50 µeq/l, resulting in Mud Pond's lower pH and elevated H^+. However, Henriksen (1982) suggests that excess SO_4^{-2} in acidic precipitation results in increased chemical mobilization of base cations, principally by increased cation exchange of H^+ for Ca^{+2} and Mg^{+2}. Thus the pre-pollution pH and alkalinity would not have been as high for either lake (Henriksen, 1982). Davis et al. (in review) suggest that the pH of Mud Pond was between 5.0 and 5.5 prior to the onset of acidic precipitation.

All of the lakes in Maine have SO_4^{-2} concentrations elevated above background (estimated at 10 µeq/l non-marine) by acidic precipitation. The amount of acidification varies in individual lakes in relation to the original buffering capacity and base cation supply of each system, and the ease with which acid was consumed by additional cation desorption and chemical weathering. Lakes with high pH (Table 6) probably have lost little or no alkalinity, whereas lakes with lower original levels of ANC may have undergone declines in alkalinity and pH.

Lakes on Granites

Table 5 lists lake water chemistry for 13 lakes located on granites in Maine, in increasing distance from the coast. Lake 1 is on Isle au Haut; lake 2 is on Mt. Desert Island; lakes 3 to 8 are in a cluster about 30 km from the coast; lakes 12 and 13 are over 250 km from the ocean. The influence of sea salts can be clearly seen as a function of distance from the coast, with Cl^- varying from 552 µeq/l offshore to 13 µeq/l well inland. Lake 8 is influenced by road salt application in its watershed, elevating the NaCl slightly. This lake illustrates the problem of identification of the existence of salt pollution near the coast and also the errors introduced by making inappropriate sea salt corrections based on Cl^- concentration.

The SO_4^{-2} concentrations, after correction for sea salts, range from 45 to 88 µeq/l. The source of the variation is largely

TABLE 6. WATER CHEMISTRY OF SELECTED LAKES ON THE SEBOOMOOK FORMATION, NORTHERN MAINE.

	Lake ID	Lake Name	pH	H^+ µeq/l	Na	Na*	K	K*	Ca	Ca*	Mg	Mg*
1	EI3-006	Glazier Lake	8.05	0	62.7	32	8.5	8	657	656	86.7	80
2	EI3-007	Wheelock Lake	7.74	0	50.7	41	4.7	4	357	356	67.8	66
3	EI3-012	Denny Pond	7.82	0	51.2	42	15.0	15	406	406	57.0	55
4	EI3-015	Round Pond	7.68	0	44.3	31	10.5	10	313	313	102	99
5	EI3-033	Roberts Pond	7.68	0	25.2	15	9.1	9	317	316	100	98
6	EI3-055	Togue Pond	7.69	0	36.4	28	8.7	8	308	308	54.7	53
7	EI3-065	Clish Pond	7.58	0	36.1	30	5.9	6	255	255	79.4	78
8	EI3-070	McClean Lake	8.35	0	52.8	40	5.9	6	1177	1176	151	148
		Average			45	32	9	8	474	473	87	85

	Lake ID	Lake Name	Cl	SO_4	SO_4*	NO_3	HCO_3	DOC mg/l	ANC µeq/l	F	ΣCations* µeq/l	$\frac{\Sigma Cat^*+Mg^*}{\Sigma Cations^*}$
1	EI3-006	Glazier Lake	36.2	93.6	90	6.5	561	6.12	629	1.4	776	0.95
2	EI3-007	Wheelock Lake	9.2	65.3	64	0.2	365	3.23	396	1.4	467	0.90
3	EI3-012	Denny Pond	10.5	112	111	0.0	374	2.55	411	5.2	518	0.89
4	EI3-015	Round Pond	14.8	92.0	91	3.3	267	7.17	312	1.2	453	0.91
5	EI3-033	Roberts Pond	12.1	77.3	76	4.2	282	8.19	317	1.0	438	0.95
6	EI3-055	Togue Pond	9.2	90.3	89	0.0	277	2.92	300	1.6	397	0.91
7	Ei3-065	Clish Pond	6.9	86.4	86	0.1	229	5.23	252	1.4	369	0.90
8	EI3-070	McClean Lake	15.4	88.9	87	2.3	1166	2.72	1297	1.0	1370	0.97
		Average	14	88	87	2	440	4.8	489	1.8	599	0.92

Sea salt corrections: Na*= Na - 0.86 Cl; K* = K - 0.02 Cl; Ca* = Ca - 0.04 Cl; Mg* = Mg - 0.2 Cl; SO4* = SO4 - 0.1 Cl

due to variation in dry deposition caused by differences in watershed vegetation and topography (Norton et al., 1988a). These values, after subtracting an estimated background SO_4^{-2} of 10 µeq/l, are equal to the maximum estimates of acidification for these lakes due to SO_4^{-2}. The ELS SO_4^{-2} median for the state is 77 µeq/l.

The values for Na^{*+}, K^{*+}, Ca^{*+2}, and Mg^{*+2} are similar and are low, typical of the weathering properties of granites in general. The SO_4^{*-2} exceeds the sum of sea salt corrected cations in only Mud Pond, which is thus acidic and acidified. Corrected for sea salt content, these lake chemistries are at or below the 1st quintile for Maine lakes (Table 4) in terms of indicated rates of chemical weathering. In Lake 11 there is a significant anion deficit and a high DOC, suggesting that organic anions are significant contributors to the total chemistry of this lake.

Lakes on the Seboomook Formation

In contrast to lakes on granites, lakes located in lithologies with carbonate such as the Seboomook Formation, a limy metasedimentary unit in northern Maine, have high concentrations of Ca^{*+2}, Mg^{*+2}, and associated HCO_3^- alkalinity (Table 6). The concentration of Ca^{+2} is close to saturation with respect to calcite for the pH. Therefore, the pH is dominated by reaction 3. Concentrations of Na^{*+} and K^{*+} for lakes located on the Seboomook Formation are not substantially different from values for the lakes on granite. The rate of chemical weathering indicated by the water chemistry of lakes from Seboomook terrain is in the fifth quintile (Table 4), approximately four times that of granites from the state.

Chloride concentrations are at background values for the state, with the exception of Glazier Lake (#1) which is anomalously high, suggesting contamination by NaCl. The SO_4^{-2} values are relatively uniform and higher than the state average of 77 by about 10 µeq/l, but not substantially higher than the regional value for that part of the state. Geologic sources may be contributing SO_4^{-2} from minor amounts present in solid solution in calcite as well as small amounts of sulfide in the bedrock and soils associated with some facies of the Seboomook Formation.

Whereas most of the waters on granite had low DOC, lake waters from Seboomook Formation terrain have higher DOC values, ranging from 2.55 to 8.19 mg/l. This higher DOC is a function of the hydrologic setting of these lakes, rather than the bedrock. There is a general relationship between DOC and the anion deficit for all the lake analyses except Lake 8. There the DOC and water color are both low, and the large anion deficit may be caused by an underestimation of HCO_3^-.

Darkwater Lakes

Darkwater lakes are defined in the ELS as having color greater than 30 Pt-Co units (Linthurst et al., 1986). Maine contained the highest percentage of darkwater lakes sampled by the ELS in the northeast. About half of the lakes had true color greater than 30 Pt-Co units. Color in most of the lakes was between 20 and 50. Only 3.4% had DOC less than 2 mg/l whereas 40.9% had DOC greater than 6 mg/l. Darkwater lakes in Maine are smaller and shallower than clearwater lakes and they have larger watersheds. Concentrations of DOC decrease as a function of increasing hydraulic residence time (Brakke et al., 1987a), suggesting that metabolism of DOC occurs in the lake. However, soil- and ground water flowpaths and residence times, and vegetation cover are probably also important factors. This relationship is commonly observed in calibrated lake studies. The association of organic anions with DOC, and the observation that the acid neutralizing capacity of lakes is related to color, suggests that at least some of the DOC is in the form of weak acids.

High Elevation Lakes

Western Maine contains most of the state's high elevation lakes. Many of these lakes are small (less than 4 ha) and consequently not represented in the ELS sample. The High Elevation Lakes in Maine (HELM) project was initiated in 1986. The project identified 90 lakes which were above 600 m, had an area greater than 1 ha, and were deeper than 1 m (Fig. 2). These lakes were sampled 1 to 3 times in 1986. Methods of sampling and analysis are detailed in Kahl (1987) and discussed by Kahl and Scott (1987), and were nearly identical to the ELS protocol. Most of the HELM lakes lie within ELS region 1C (Fig. 1). Although the two surveys were conducted two years apart, comparison is useful in order to evaluate the influence of the different environmental variables identified above. Four lakes sampled by ELS in 1984 and by HELM in 1986 had nearly identical chemistry in both years.

The HELM lakes are more acidic (Fig. 10), have lower ANC (Fig. 11), lower Ca^{*+2} and Mg^{*+2} and Cl^- (Table 7), and higher SO_4^{*-2} (Fig. 12) than the ELS lakes. HELM lakes, as a group, are located on non-calcareous geologic substrates, probably have thinner acidic soils with lower base saturation, and perhaps shorter water residence times, all of which render them more sensitive to acidic inputs. The median SO_4^{-2} in HELM lakes is 3 times the SO_4^{-2} in precipitation. The higher concentrations are probably caused by higher loading rates associated with cloud and fog interception, as observed in other mountainous areas in the northeast (Lovett et al., 1982). The combination of high sensitivity to acidification and higher SO_4^{-2} loading results in the lakes being more acidic, with a larger proportion of acidic lakes. In 1986, 5% of the HELM lakes had a pH less than 5, 13% had ANC less than 0, and 70% had SO_4^{-2} greater than 80 μeq/l. The corresponding figures for ELS lakes are 0.6%, 0.6%, and 40%, respectively. Studies of the biological and chemical evidence from sediment cores from several of these high elevation lakes has suggested that they have become acidified over the last 100 years, probably because of atmospheric deposition of SO_4^{-2} (Davis et al., 1983; Charles and Norton, 1986).

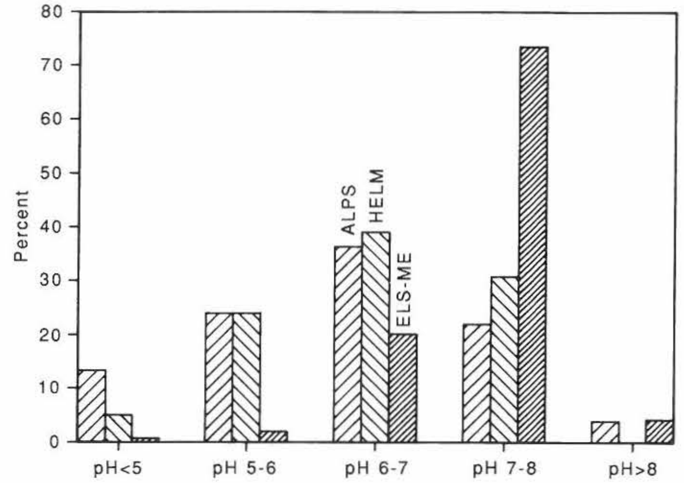

Figure 10. Air-equilibrated pH distribution for ELS, HELM, and ALPS lakes in Maine.

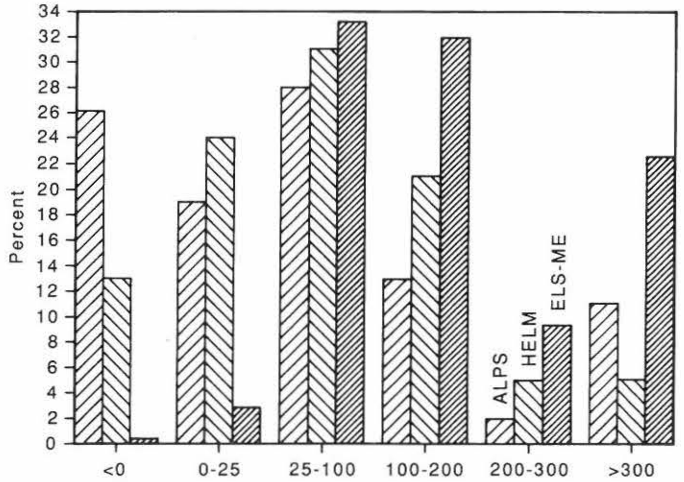

Figure 11. Gran plot ANC distribution for ELS, HELM, and ALPS lakes in Maine. ANC in μeq/l.

Aquifer Lakes

A seepage lake was defined for the ELS as having no surface inlet or outlet. However, many lakes located in stratified sand and gravel deposits (kames, eskers, and deltas) have outlets, and were thus classified as drainage lakes by ELS regardless of the source of their hydrologic and chemical inputs, which are seepage in nature. We define the Maine population of aquifer lakes as those with no inlets, and thus limit them to lakes receiving inputs only from seepage and from the atmosphere. Most of the aquifer lakes are small and thus under-represented in the ELS population. We have identified approximately 120 such lakes with areas greater than 0.4 ha and depths of at least 1 m. Eighty-nine lakes were sampled in the fall of 1986 (Aquifer Lake

TABLE 7. COMPARATIVE CHEMISTRY FOR SAMPLED ELS, HELM, AND SEEPAGE (AQUIFER) LAKES IN MAINE.

	n	N	Mean air pH	Median air pH	Mean ANC	Median ANC	Mean SO_4*	Median SO_4*	Mean SO_4	Median SO_4	Median Cl	Median Sum of Ca*+Mg*+Na*+K*
HELM	91	91	5.73	6.71	82	48	84	85	85	86	10	155
Aquifer	128	140	5.15	6.53	136	40	44	38	53	46	48	84
ELS	225	1966	6.47	7.26	186	129	71	70	76	76	27	226

N = the estimated total population; n = the number of sampled lakes; Values in μeq/l except pH. * = corrected for marine aerosol content.

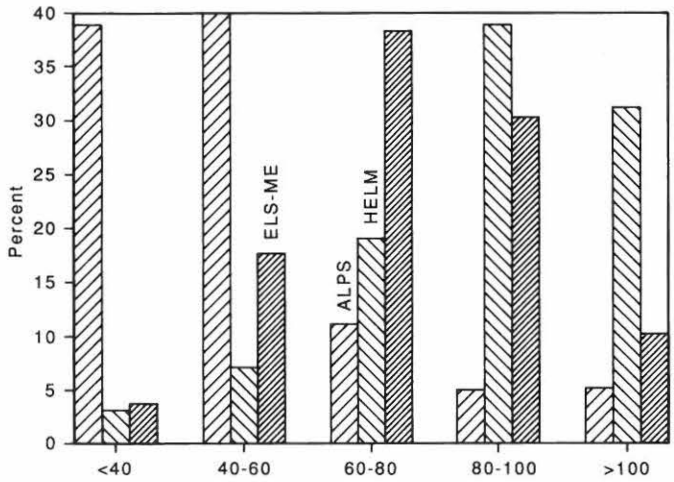

Figure 12. Sulfate distribution for ELS, HELM, and ALPS lakes in Maine. Sulfate in μeq/l.

Project; Fig. 2). Collection and analytical methods were the same as for HELM.

As a group, these lakes lie at lower elevations than the ELS lakes, ranging from 50 to 200m, and their geographic distribution is biased toward the coastal half of the state (Fig. 2). This distribution imparts a regional bias to such environmental parameters as dry deposition of marine aerosols. Additionally, the likelihood of disturbance of the watersheds, including deforestation, construction, or agriculture, is increased because of higher development in this part of the state. Watersheds of several aquifer lakes contained domestic blueberry fields.

Of the three groups of lakes for which data are shown in Figures 10 to 12, seepage or aquifer lakes are the most acidic, have the lowest ANC, Ca^{*+2}, Mg^{*+2}, DOC, and SO_4^{*-2} concentrations and, in spite of the low pH, also have low dissolved Al due to the relative lack of contact between acidic waters and watershed soils. Thirteen percent of the aquifer lakes had pH values below 5, 26% had ANC less than 0, and only 10% had SO_4^{-2} concentrations greater than 80 μeq/l in spite of having greater sea salt content and lower pH than the HELM and ELS lakes. The corresponding figures for the ELS lakes are 0.6%, 0.6%, and 40%, respectively. These seepage lakes have very small watersheds, as indicated by the low concentrations of Ca^{*+2} and Mg^{*+2} which are derived from soil weathering processes. Low contribution of terrestrial alkalinity generation to the lakes renders the aquifer lakes sensitive to acid inputs because of lower pre-pollution alkalinity. The average SO_4^{-2} concentration is about 25 μeq/l lower than the ELS mean. We suggest that these SO_4^{-2} values are derived as follows. Precipitation in Maine has SO_4^{-2} concentrations of about 30 μeq/l (NADP, 1986). The SO_4^{-2} becomes concentrated by evapotranspiration by a factor of about 1.6X, yielding 50 μeq/l, the value in these lakes. The difference between the ELS lake median SO_4^{-2} (77 μeq/l) and aquifer lake median SO_4^{-2} (47 μeq/l) is attributed to a lack of dry deposition to aquifer lakes. The difference between the median SO_4^{-2} for aquifer and HELM lakes is about 40 μeq/l. Thus vegetational effects, combined with cloud and fog interception, produce nearly a doubling of the SO_4^{-2} in lakes over the expected concentrations from typical wet precipitation.

SUMMARY

The major ion chemistry of lakes in Maine is controlled by a number of factors. These include the bedrock geology and soil composition, wet precipitation chemistry, chemistry of dry-deposited gases and aerosols, hydrology, watershed vegetation, and land use. For lakes in similar geologic terrain such as granites, surface water chemistry varies markedly within the state due to pronounced gradients in the contribution of marine aerosol and in wet and dry deposition of acidic substances, especially SO_4^{-2}. The rate of chemical weathering varies widely, with a variation of over 500 percent at the first and fourth quintile. With the exception of road salt pollution and excess SO_4^{-2} from acidic precipitation, the major ion chemistry of lowland Maine lakes is controlled by natural processes, and the typical lake is circumneutral. The contribution of anthropogenic SO_4^{-2} to the acidity status of Maine lakes ranges from negligible to dominant.

High elevation lakes and aquifer (seepage) lakes appear to be influenced by acidic precipitation, although many of the acidic lakes may also be strongly influenced by natural organic acidity. These two groups of lakes contain most of the acidic lakes in the state.

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