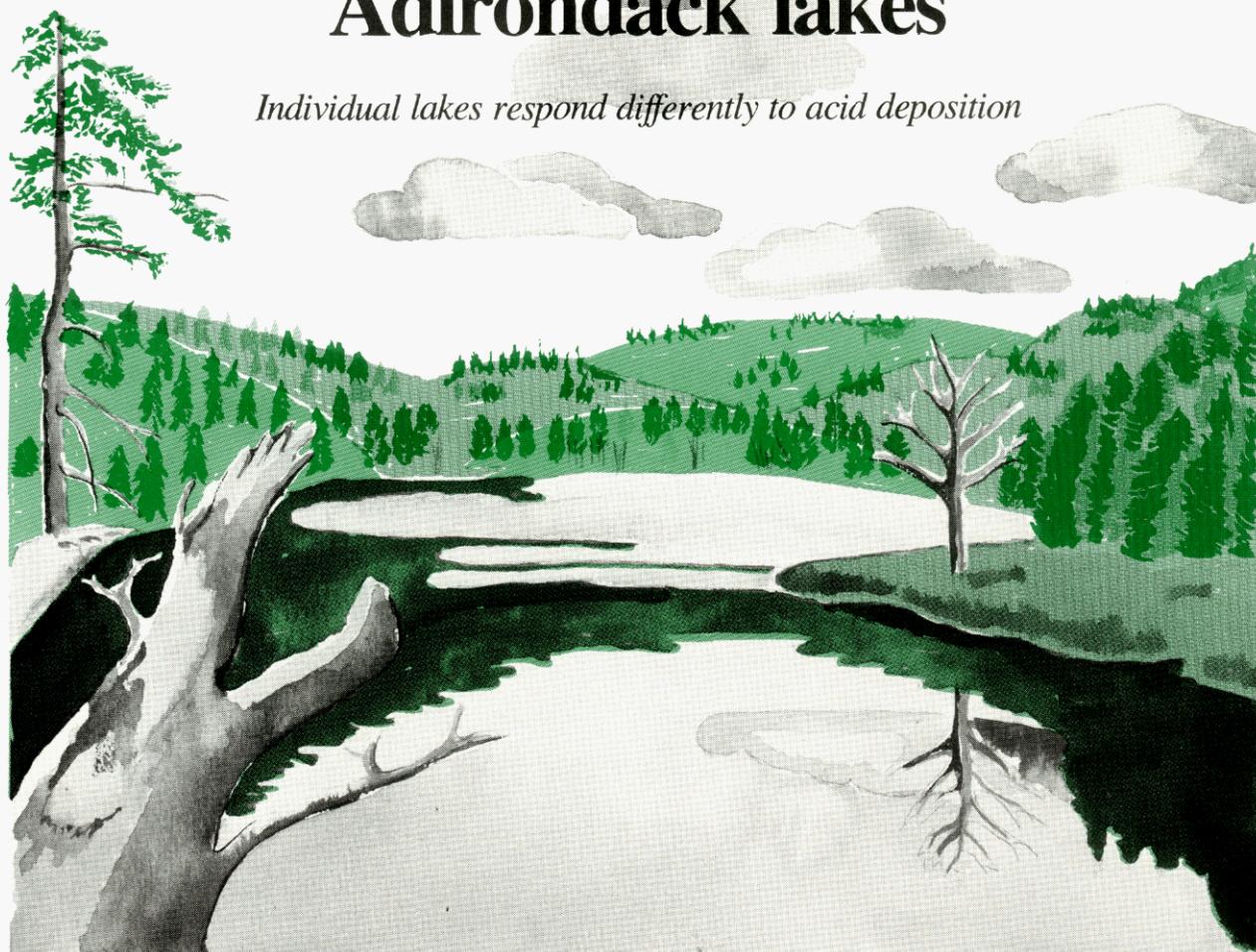


Chemical characteristics of Adirondack lakes

Individual lakes respond differently to acid deposition



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There is much concern over the role of atmospheric deposition of mineral acids in the acidification of low-ionic-strength (dilute) surface waters in remote regions. Surface water acidification has been attributed to atmospheric deposition of H_2SO_4 (or SO_2) (1) and HNO_3 (2), the oxidation of organic nitrogen from soil, the production of soluble organic acids through the decay of

dead plant and animal material in soil (3), the oxidation of naturally occurring sulfide minerals (4), and changes in land use (5, 6).

In the northeastern U.S., many areas are sensitive to mineral acidity. Acid-sensitive watersheds are generally underlain by granitic bedrock and have small pools of readily available (exchangeable or easily weatherable) basic cations (C_B ; Ca^{2+} , Mg^{2+} , Na^+ , K^+).

These watersheds are also characterized by an inability to retain inputs of acidic anions (SO_4^{2-} , NO_3^- , Cl^-). That is, atmospheric inputs are transported through the watershed, rather than being retained. Therefore, high loadings of acidic anions will not be attenuated

in the soil but will be removed from the watershed with drainage water. Anionic solutes must be accompanied by an equivalent charge of cations to maintain electroneutrality (charge balance). Complete neutralization of acidity can be accomplished by the dissolution or exchange of basic cations within the soil.

In the absence of readily available pools of basic cations, however, neutralization will be incomplete, and acidic cations— H^+ , labile monomeric Al (Al^{n+})—will be transported from the soil to the surface water (7). The effects of this process cause concern because high concentrations of acidic cations appear to be harmful to aquatic biota

and surface water quality (8–11).

The research reported here was conducted as part of the Regionalized Integrated Lake–Watershed Acidification Study (RILWAS), the objectives of which are discussed in detail elsewhere (12). Our intent was to evaluate the general chemical characteristics of lakes in the Adirondack region of New York and to assess the mechanisms that regulate the acid–base chemistry of these waters.

Study sites and methods

Twenty lakes and watersheds in the Adirondacks were sampled for a period of 26 months at approximately monthly intervals (Figure 1; Table 1). More intensive sampling was done during spring snowmelt. Samples were collected from drainage lake outlets and seepage lake surfaces. In an effort to evaluate mechanisms that control the sensitivity of Adirondack lakes to acidification, the study lakes were selected to include a range of geologic and hydrologic characteristics (Table 1).

The analytical methods used in this study are summarized in Table 2. Samples were measured for temperature and pH and extracted for monomeric Al in the field to minimize transformations (e.g., changes in temperature and degassing of CO₂) that might alter the measurements of pH and Al. Samples were refrigerated after collection and transported to a laboratory at Syracuse University. There they were analyzed for dissolved inorganic carbon (DIC) and nitrogen forms, placed in ampules for the determination of dissolved organic carbon (DOC), and processed for the determination of nonlabile monomeric Al. These samples were analyzed as soon after collection as possible, generally within 72 h. Samples were stored at 4 °C until all analytical determinations had been made.

The distribution of DIC and labile monomeric (inorganic) Al was calculated using a chemical equilibrium model. The equilibrium constants and enthalpy values used for this analysis are summarized elsewhere (23). Calculations were corrected for the effects of ionic strength, using the Debye–Hückel approximation for individual ion activity coefficients (24), and temperature.

Charge balance calculations were made on all samples. Cationic equivalence (charge) was assumed to be distributed among basic cations (Ca²⁺, Mg²⁺, Na⁺, K⁺), NH⁴⁺, H⁺, and Alⁿ⁺. The charge equivalence of Alⁿ⁺ was calculated by determining OH[−], F[−], and SO₄^{2−} complexes, using a chemical equilibrium model. Anionic equivalence was distributed among acidic anions (SO₄^{2−}, NO₃[−], Cl[−]), free F[−], HCO₃[−], and free organic anions

(RCOO[−]). The amount of bicarbonate was calculated from DIC, pH, and temperature measurements, using the equilibrium model. The concentration of free organic anions was calculated as the difference between the charge of inorganic cations and the charge of inorganic anions. To allow a comparison between labile (inorganic) and nonlabile (organic) forms of monomeric Al, nonlabile monomeric Al was assumed to be trivalent and bound by an equivalent organic ligand.

Chemical characteristics

The Adirondack region of New York State receives substantial atmospheric deposition of mineral acids (0.5–0.75 kiloequivalents [keq]/ha·y SO₄^{2−}; 0.25–0.4 keq/ha·y NO₃[−]) (25). Soils within the region are generally Spodosols, which adsorb minimal quantities of SO₄^{2−} (26). Sulfate in surface waters apparently originates largely from atmospheric deposition (27, 28).

Although much of the region is underlain by acid-sensitive bedrock, the chemistry of surface waters varies widely from about neutral to acidic (29, 30). These variations are the result of the geologic and hydrologic characteristics of the different watersheds (31,

32). In some of the watersheds, natural sources of acidity significantly influence the acid–base chemistry of surface waters.

Average values of relevant chemical characteristics for the 20 lakes in this study are summarized in Table 3. There is a wide range of pH, acid-neutralizing capacity (ANC), NO₃[−], Ca²⁺, Al, and DOC in the lakes. Sulfate is the dominant anion in all lakes, except for the relatively insensitive Black and Woodruff ponds, in which HCO₃[−] is the major anion. There is a relatively narrow range of SO₄^{2−} concentrations among Adirondack drainage lakes (106–154 μeq/L; this range does not include Barnes Lake and Little Echo Pond, which are seepage lakes).

We have selected a subset of five lakes to illustrate the range in chemical compositions of Adirondack waters (Figure 2). A study of the chemistry of these lakes, together with the geology and hydrology of their watersheds, allows us to make an evaluation of the significant processes controlling sensitivity to acid deposition.

Seepage lakes

Seepage lakes are those that have no surface inlets or outlets. They are gen-

FIGURE 1
Adirondack lakes in the study^a

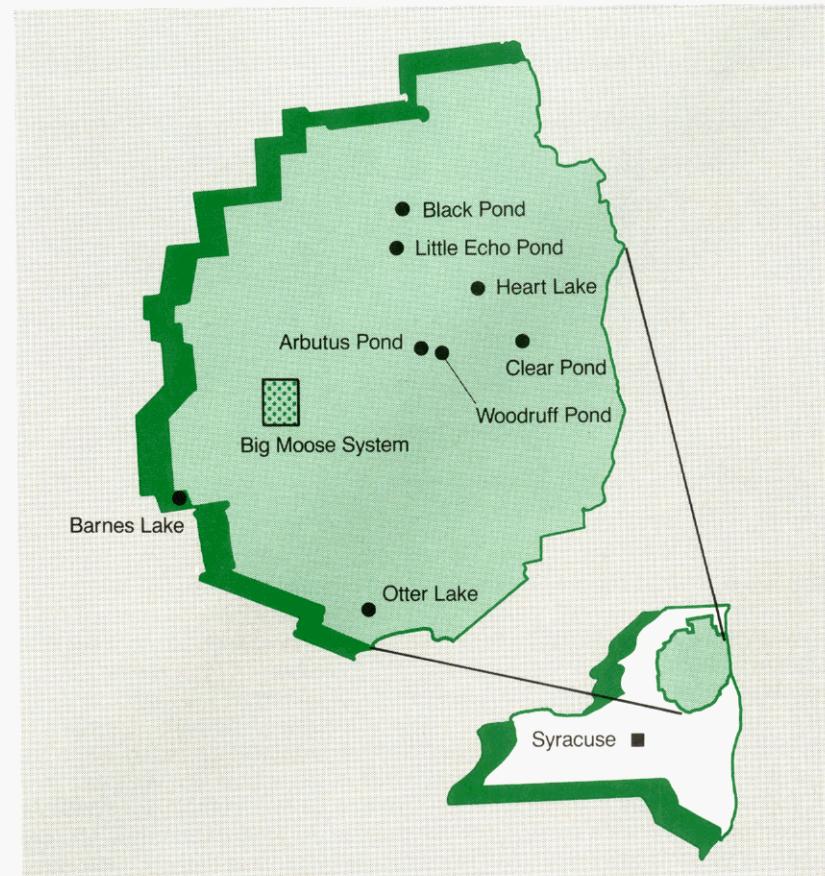


TABLE 1
Location and physical characteristics of Adirondack study lakes and their watersheds

Lake	Location	Elevation (m)	Lake type	Surficial geology	Maximum depth (m)	Surface area (ha)	Volume (10^4 m^3)
Arbutus Pond	43°58'N, 74°14'W	513	Drainage	Thin/thick till	8.4	50	146
Barnes Lake	43°34'N, 75°14'W	397	Seepage	Glacial sand	9.0	5.1	7.7
Big Moose Lake	43°49'N, 74°53'W	557	Chain drainage	Thin till	22.0	520	3600
Black Pond	44°26'N, 74°18'W	494	Drainage	Thick till/stratified drift	14.0	32	199
Bubb Lake	43°57'N, 74°51'W	554	Chain drainage	Thin till	3.0	20	42
Cascade Lake	43°48'N, 74°53'W	554	Drainage	Thin/thick till	5.0	40	172
Clear Pond	43°59'N, 74°50'W	582	Drainage	Thin/thick till	24.0	73	660
Constable Pond	43°49'N, 74°50'W	584	Chain drainage	Thin till	4.0	23	45
Darts Lake	43°48'N, 74°53'W	536	Chain drainage	Thin till	16.0	144	415
Heart Lake	44°26'N, 74°18'W	662	Drainage	Thin/thick till	13.0	11	54
Little Echo Pond	44°18'N, 74°22'W	481	Seepage	Sphagnum bog	5.0	0.8	2.8
Merriam Lake	43°51'N, 74°51'W	651	Drainage	Thin till	4.9	7.8	11.5
Moss Lake	43°46'N, 74°51'W	536	Chain drainage	Thin/thick till	15.0	45	272
Otter Lake	43°11'N, 74°30'W	488	Chain drainage	Thin till	4.5	10	22
Lake Rondaxe	43°11'N, 74°55'W	525	Chain drainage	Thin/thick till	10.0	92	220
Squash Pond	43°48'N, 74°51'W	650	Drainage	Thin till	7.0	3.9	7.2
Townsend Pond	43°48'N, 74°51'W	561	Drainage	Thin till	—	—	—
West Pond	43°48'N, 74°52'W	579	Drainage	Peat/thin till	4.0	12	—
Windfall Pond	43°48'N, 74°51'W	595	Drainage	Thin till	5.5	1.5	4.9
Woodruff Pond	43°48'N, 74°10'W	470	Drainage	Carbonate; thin/thick till	3.4	18.1	34

TABLE 2
Chemical characteristics analyzed in the study

Characteristics	Methods	Reference
SO_4^{2-} , NO_3^- , Cl^-	Ion chromatography	13
NH_4^+	Phenate colorimetry; autoanalysis	14
Ca^{2+} , Mg^{2+} , Na^{2+} , K^+	Atomic absorption spectroscopy (AAS)	15
pH	Potentiometric with glass electrode; field measurement	16
Acid neutralizing capacity (ANC)	Strong acid titration with Gran plot analysis	17
Dissolved inorganic carbon (DIC)	Syringe stripping of CO_2 ; gas chromatography	18
Dissolved organic carbon (DOC)	Filtration; persulfate oxidation; syringe stripping of CO_2 ; gas chromatography	19
Dissolved silica	Heteropoly blue complex colorimetry; autoanalysis	16
Total F	Potentiometric with ion-selective electrode after addition of total ionic strength adjustment buffer	20
Monomeric Al	Field extraction by 8-hydroxy quinoline into methylisobutyl ketone; analysis by AAS; graphite furnace atomic absorption	21, 22
Nonlabile monomeric Al	Fractionation by ion exchange column; analysis for monomeric Al	22
Specific conductance	Conductivity bridge	16
Temperature	Thermometer	—

erally found in areas covered by highly permeable glaciofluvial sand and gravel. The two seepage lakes in this study are isolated from the surrounding groundwater systems. Barnes Lake is a perched clearwater seepage lake isolated above the regional water table by organic-rich bottom sediments (Figure 3a). Little Echo Pond is a brownwater seepage lake surrounded by thick (~8 m) peat deposits (Figure 3b). The low permeability of the peat prevents significant movement of groundwater into this lake.

The concentrations of C_B , Al, and dissolved Si are low in both lakes (Figure 2). These low concentrations and the hydrology of the two lakes are indications that water enters primarily from precipitation. The lack of surface runoff or groundwater in the lakes is the reason the waters are deficient in dissolved Si and contain low concentrations of Al, despite their low pH. Lakes that receive most of their water directly from precipitation are extremely sensitive to atmospheric deposition.

Although both lakes are acidic, there

are significant differences in their chemical characteristics. The dominant anion in Barnes Lake is SO_4^{2-} ; in Little Echo Pond there appear to be comparable concentrations of SO_4^{2-} and organic anions (on an equivalence basis), demonstrated by the large discrepancy in charge balance. This relatively large concentration of organic anions is consistent with the high concentrations of DOC in Little Echo Pond. Although much of the acidity of the pond can be attributed to atmospheric deposition of H_2SO_4 , it appears that the peat deposits and the sphagnum mat surrounding the pond release substantial quantities of naturally occurring organic acids. These substances then dissociate and contribute to overall acidity.

Drainage lakes

The chemistry of drainage lakes, for example, Merriam Lake, West Pond, and Cascade Lake (Figure 2), is considerably different from that of seepage lakes. Drainage lakes have outlet streams and receive water by stream discharge and seepage, as well as by direct atmospheric deposition. The higher ionic strength and higher concentrations of C_B and dissolved Si are the result of contact between the drainage water and the soil (Figure 4).

Merriam Lake contains highly acidic drainage water. It is a headwater lake into which water drains from very shallow (<1-m deep) acidic soil. Because of a lack of readily available C_B , the neutralization of mineral acid deposited in this catchment is accomplished in part by the dissolution of Al in the soil. Consequently, SO_4^{2-} is the dominant anion and H^+ and Aln^+ are found in significant concentrations.

TABLE 3
Chemical characteristics of Adirondack study lakes^a

Lake	pH	ANC (μeq/L)	NO ₃ ⁻ (μeq/L)	SO ₄ ²⁻ (μeq/L)	Ca ²⁺ (μeq/L)	Monomeric Al (μmol/L)	DOC (μmol/L)
Arbutus Pond	6.2 ± 0.3	58 ± 21	10 ± 6	141 ± 10	152 ± 11	1.0 ± 0.6	420 ± 65
Barnes Lake ^b	4.7 ± 0.1	-14 ± 10	2 ± 3	65 ± 12	30 ± 4	1.1 ± 0.6	450 ± 136
Big Moose Lake	5.1 ± 0.1	1 ± 10	24 ± 5	140 ± 11	93 ± 10	8.9 ± 2.7	340 ± 78
Black Pond	6.8 ± 0.2	200 ± 21	4 ± 5	130 ± 3	191 ± 11	0.3 ± 0.6	350 ± 65
Bubb Lake	6.1 ± 0.2	41 ± 28	16 ± 7	131 ± 14	108 ± 13	1.8 ± 1.3	280 ± 82
Cascade Lake	6.5 ± 0.3	95 ± 52	29 ± 9	139 ± 10	159 ± 24	2.8 ± 1.9	310 ± 92
Clear Pond	7.0 ± 0.2	100 ± 19	1 ± 2	139 ± 11	157 ± 21	0.8 ± 0.7	320 ± 64
Constable Pond	5.2 ± 0.6	8 ± 22	17 ± 12	149 ± 19	98 ± 10	10.5 ± 4.3	420 ± 80
Darts Lake	5.2 ± 0.2	6 ± 12	24 ± 5	139 ± 7	97 ± 9	7.6 ± 2.7	320 ± 86
Heart Lake	6.4 ± 0.3	43 ± 12	5 ± 6	106 ± 11	119 ± 12	0.6 ± 0.6	310 ± 51
Little Echo Pond ^b	4.3 ± 0.1	-51 ± 17	0 ± 0	78 ± 17	36 ± 6	1.2 ± 0.5	1100 ± 153
Merriam Lake	4.5 ± 0.2	-25 ± 15	21 ± 13	137 ± 11	58 ± 7	19.0 ± 0.6	480 ± 110
Moss Lake	6.4 ± 0.2	66 ± 25	26 ± 6	141 ± 8	146 ± 15	2.2 ± 1.6	310 ± 61
Otter Lake	5.5 ± 0.5	13 ± 16	9 ± 9	138 ± 21	88 ± 11	5.0 ± 3.6	200 ± 54
Lake Rondaxe	5.9 ± 0.5	33 ± 25	23 ± 6	134 ± 7	112 ± 10	4.4 ± 3.0	300 ± 60
Squash Pond	4.6 ± 0.6	-22 ± 39	24 ± 17	131 ± 18	65 ± 28	19.2 ± 7.6	580 ± 127
Townsend Pond	5.2 ± 0.6	18 ± 21	27 ± 15	154 ± 30	95 ± 16	9.9 ± 7.9	260 ± 81
West Pond	5.2 ± 0.5	29 ± 50	10 ± 6	111 ± 13	94 ± 24	6.6 ± 2.1	670 ± 204
Windfall Pond	5.9 ± 0.4	44 ± 30	26 ± 14	141 ± 17	143 ± 19	5.6 ± 2.4	390 ± 92
Woodruff Pond	6.9 ± 0.2	410 ± 140	2 ± 3	147 ± 17	430 ± 120	1.0 ± 1.1	710 ± 161

^a Arithmetic mean and standard deviations shown for the samples collected

^b Barnes and Little Echo are seepage lakes; the others are drainage lakes

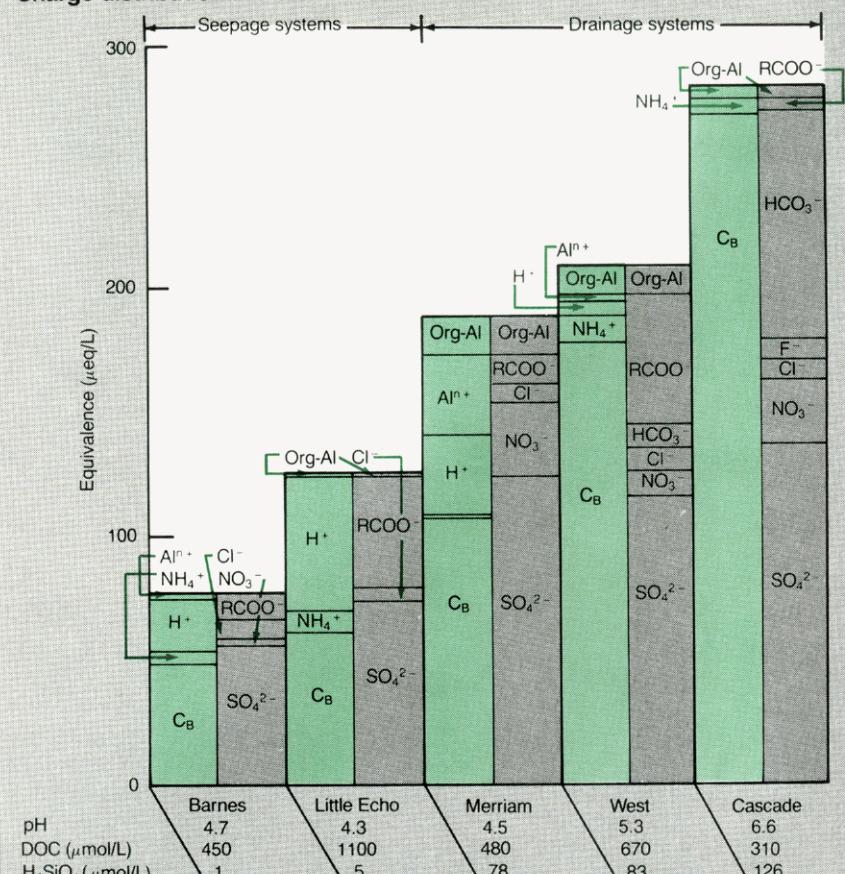
Although SO₄²⁻ is the predominant anion in Merriam Lake waters, elevated concentrations of NO₃⁻ and, to a lesser extent, organic anions also may contribute to the acidity. The modest concentrations of DOC, organic anions, and nonlabile monomeric Al probably are indicators of the importance of forest floor mineralization processes in the shallow acidic soils within the watershed.

Nitrate represents a significant source of acidity in many Adirondack drainage lakes (33). Moreover, short-term increases in H⁺ and Alⁿ⁺ that occur in chronically acidic lakes during hydrologic events are associated with NO₃⁻ inputs (33). The source of the NO₃⁻ is unclear, but it may be the result of snow or seasonal variations in the mineralization and assimilation of nitrogen by terrestrial biota and microorganisms.

The chemistry of Merriam Lake is characteristic of chronically acidic lakes. In these lakes, the equivalence of SO₄²⁻ exceeds that of C_B and NH⁺. This difference suggests that H⁺ and Alⁿ⁺ acidity is largely the result of sulfuric acid deposition. Aluminum in clearwater acidic lakes such as Merriam occurs primarily in an inorganic (labile) form (aquo Al and Al in OH⁻, F⁻, and SO₄²⁻ complexes), which is thought to be toxic to fish (10, 34, 35) and other aquatic life (36).

West Pond is a bog drainage system with thick deposits of peat that cover a large area immediately adjacent to the pond. Although the pond is characteris-

FIGURE 2
Charge distribution in Adirondack lake waters^a



^a Values represent 26-month averages
Barnes and Little Echo are clearwater and brownwater seepage ponds, respectively. Merriam, West, and Cascade are acidic, bog, and neutral pH drainage lakes, respectively.

C_B is the sum of basic cation (Ca²⁺, Mg²⁺, Na⁺, K⁺) equivalence

The equivalence of labile monomeric Al (Alⁿ⁺) is calculated by considering the various inorganic complexes

Non-labile monomeric Al (Org-Al) is assumed to be trivalent and balanced by an equivalent organic ligand

Free organic anions are calculated as the difference between inorganic cations and inorganic anions

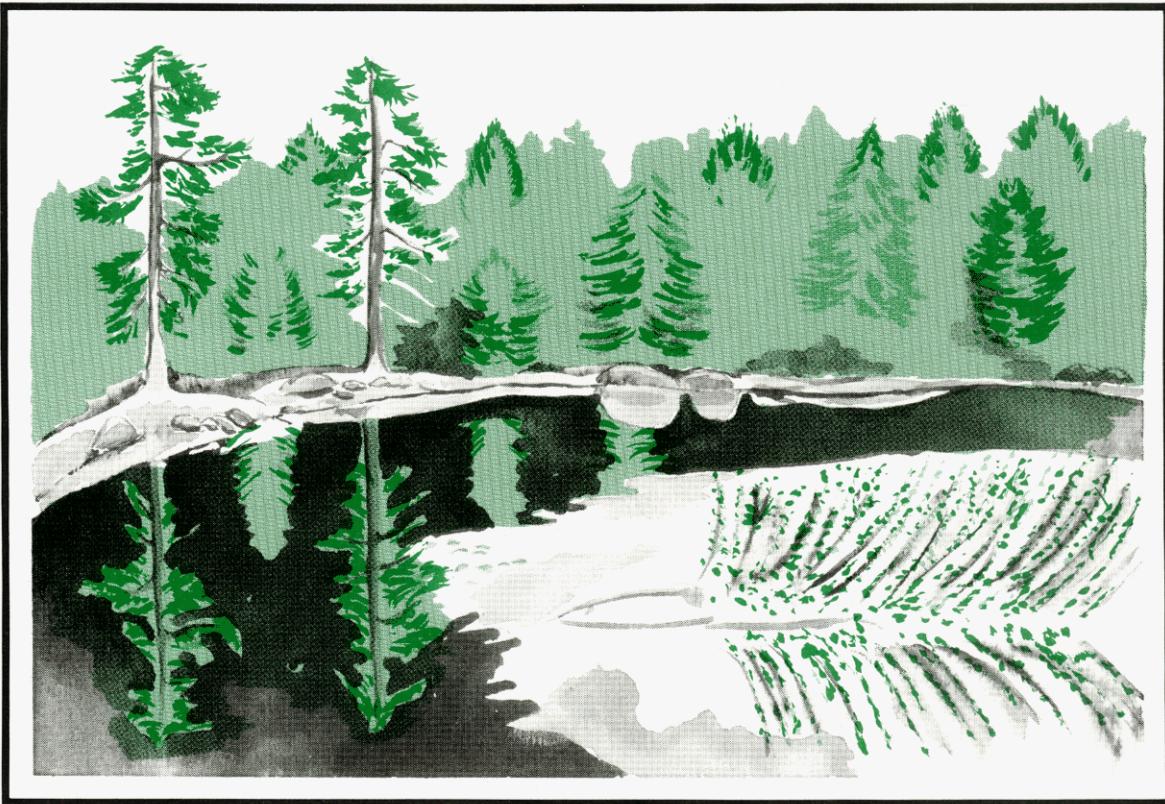
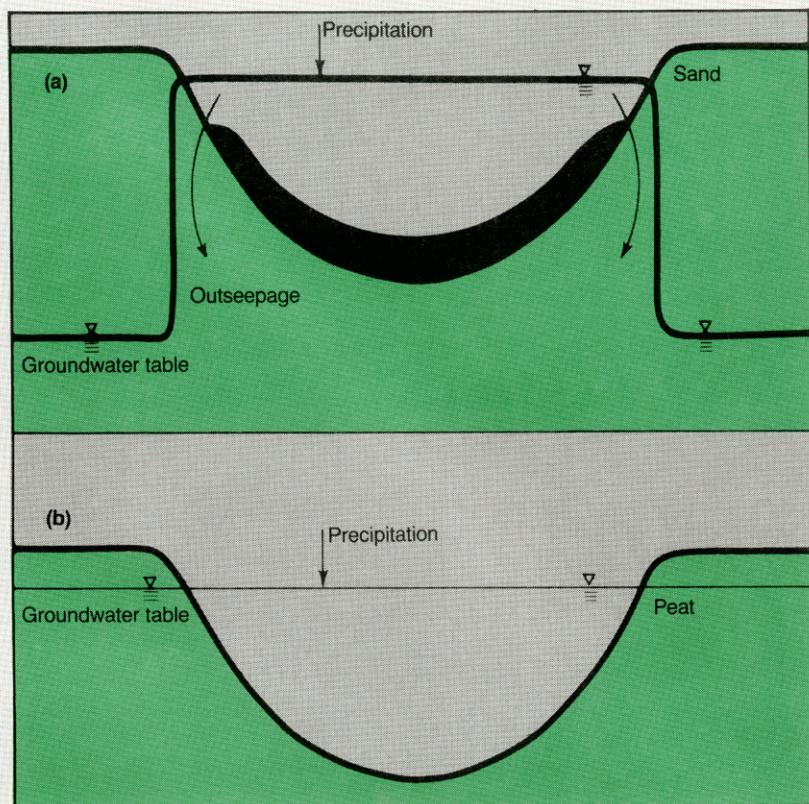


FIGURE 3
Acid-sensitive seepage lakes^a



^aSurface water in these lakes is isolated from the surrounding groundwater. In perched seepage lakes (a) the water is above the regional groundwater table because of the low permeability of organic rich bottom sediments. Bog seepage lakes (b) are surrounded by thick peat deposits that restrict significant movement of groundwater into the lake. Solute concentrations in isolated seepage lakes are strongly influenced by atmospheric deposition.

tically acidic, unlike clearwater acidic lakes its acidity is attributed to sulfuric and organic acids alike. Because West Pond is surrounded by thick organic peat deposits, most of the Al appears to be associated with organic matter (in the nonlabile monomeric form) and therefore is probably less toxic to aquatic organisms than the labile inorganic form of Al (10, 35).

Cascade Lake is generally illustrative of Adirondack waters that are relatively insensitive to atmospheric deposition of mineral acids. Although there are significant concentrations of SO_4^{2-} in the drainage waters, deposition of acids appears to be entirely compensated for by the dissolution of C_B from the soil. The high concentrations of HCO_3^- and dissolved Si indicate that the extent of weathering reactions is greater there than in more acidic lake systems. In part, these trends are the result of thick till deposits (that is, soil and gravel deposited by prehistoric glaciation) within the catchment area that allow the waters that move through the watershed to have greater contact with the mineral soil (Table 1).

The approximately neutral pH values and low DOC concentrations in Cascade Lake are consistent with the low concentrations of monomeric Al. The low concentrations of Al appear to be associated largely with organic solutes (in a nonlabile monomeric form).

Differences in lake chemistry

It is evident that there are profound lake-to-lake variations in sensitivity to

acidification and acidification processes. The southern and western Adirondacks receive the highest loading of SO_4^{2-} , whereas the northeastern section receives less (25). Although there are differences in the atmospheric loading of SO_4^{2-} in the Adirondacks, these intraregional variations appear minor, as evidenced by the uniform SO_4^{2-} concentrations found in drainage waters (Table 3).

Differences in lake sensitivity and water chemistry are largely the result of variations in water flow paths and in the contact of water with the surrounding surficial geology (Figures 3 and 4) (31, 32). For example, the presence of thick peat deposits can have a profound effect on water chemistry. Waters that come into contact with such deposits are rich in organic acids, which contribute to acidity and bind potentially toxic forms of Al. The brownwater lakes are probably naturally acidic, although deposition of SO_4^{2-} undoubtedly has enhanced their acidity (e.g., Little Echo and West ponds; Figure 2).

Clearwater lakes show a distinct range in sensitivity to acid deposition. This sensitivity is a function of flow paths of water through catchments or of the presence of weatherable minerals (e.g., calcite) or both. Seepage lakes isolated from the local water table (e.g., Barnes Lake) are extremely sensitive to strong acids because of the minimal contact of water with the mineral soil and therefore the minimal neutralization of acidity from the release of C_B (Figure 3).

Waters that drain from shallow acidic soils also are sensitive to mineral acids. Pools of exchangeable or easily weatherable C_B in the soil are small, and the retention time of water in the mineral soil is short (32). Therefore, H^+ neutralization is incomplete and results in the dissolution of Al^{n+} and the transport of H^+ and Al^{n+} to surface waters (Figure 4).

If there is adequate contact of drainage waters with deeper mineral soil—thick till, for example—or an abundance of easily weatherable minerals, such as carbonate, C_B will be released to compensate for mineral acids and will neutralize CO_2 acidity in the soil. These processes result in completely neutralized mineral acids and produce HCO_3^- in solution (Figure 4).

The sensitivity of surface water to acidification largely depends on the extent to which C_B is released relative to the deposition and retention of acidic anions. In the Adirondacks, as the equivalents of SO_4^{2-} and NO_3^- in solution approach and exceed the equivalents of C_B , neutralization of mineral acids is incomplete, and elevated concentrations of potentially harmful

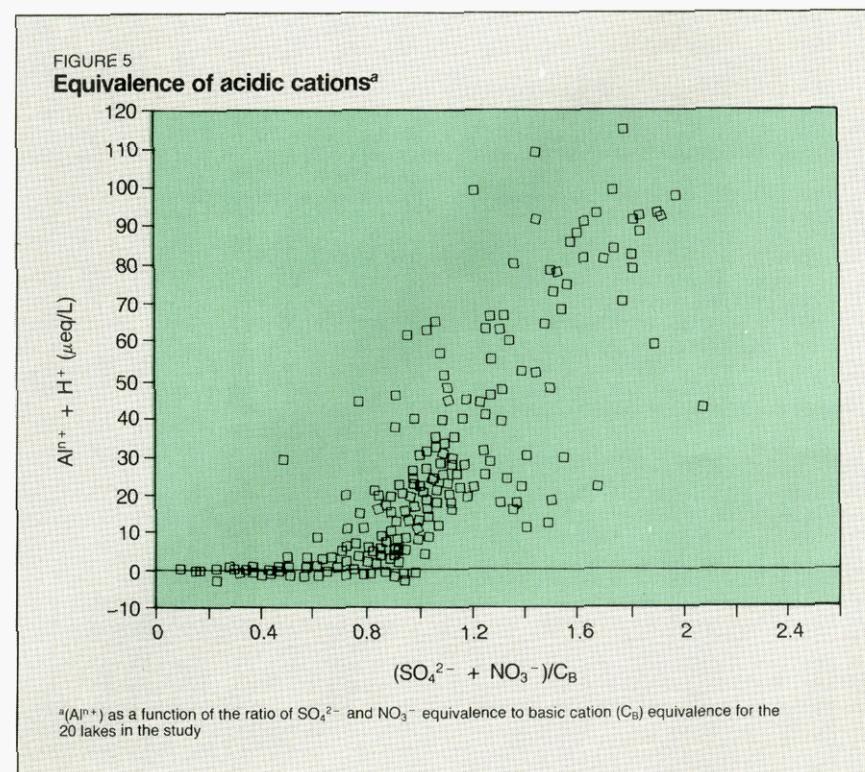
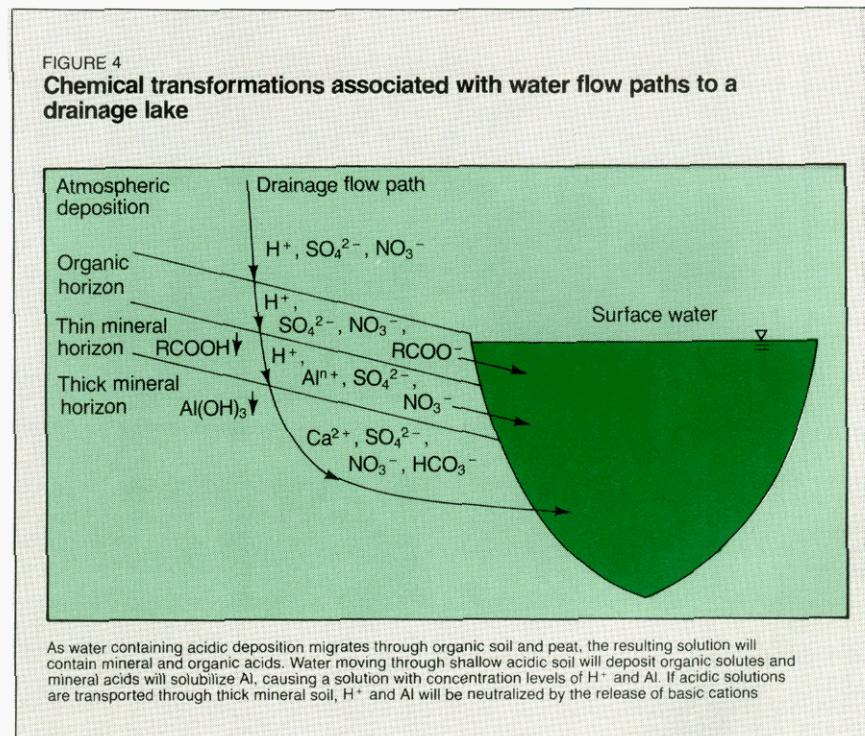
acidic cations (H^+ , Al^{n+}) are evident in surface waters (Figure 5). Although organic acids are the main cause of brownwater lake acidity, H_2SO_4 and, to a lesser extent, HNO_3 are predominantly responsible for the acidification of clearwater Adirondack lakes.

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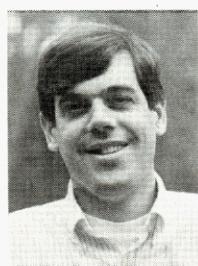




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