

Historical Changes in Connecticut Lakes Over a 55-Year Period

Peter A. Siver,* Richard W. Canavan IV, Cathryn K. Field, Laurence J. Marsicano, and Anne-Marie Lott

ABSTRACT

Changes in the chemical and physical conditions of 42 Connecticut lakes are compared between three time periods, the late 1930s, the mid- to late 1970s and the early 1990s. On average, lakes have decreased in Secchi disk depth by 1.2 m and doubled in total phosphorus concentration, many in a unidirectional manner. As a result, the suite of lakes can be characterized as having shifted from an oligo-mesotrophic condition (1930s) to a late mesotrophic condition (1990s). Since the 1970s, lakes have increased in base cation concentrations an average of 70 $\mu\text{eq/L}$, many as the result of an increase in sodium. Increases in sodium were generally coupled with increases in chloride ions. Many of the lakes positioned in watersheds that have become more residential since the 1930s and/or 1970s have also increased in alkalinity. Despite the overall increase in base cations, chloride, and alkalinity, about 25% of the waterbodies that have remained situated in primarily forested watersheds in crystalline rock regions have decreased in total cation concentrations; about half of these lakes have also significantly decreased in alkalinity since the 1930s. The changes are discussed in relation to the degree of urbanization of the watersheds over the same time period.

IT IS RARE to find comparative historical chemical and physical data on lakes as old as 50 yr (e.g., Eilers et al., 1989; Brenner et al., 1993). As a result of the paucity of historical data for most regions, paleolimnological techniques have been used to reconstruct past lake water conditions (Smol, 1992). Likewise, it is equally rare to have 50-yr-old records of land uses in lake watersheds. We are fortunate in Connecticut to have 55-yr-old chemical and physical data on many lakes in the state, and aerial photographs from the same time periods from which watershed land use can be evaluated.

Between 1937 to 1939, Deevey (1940) surveyed chemical and physical characteristics of 46 Connecticut waterbodies. In particular, Deevey made measurements of Secchi disk depth, total phosphorus concentrations and alkalinity. In 1934, the State of Connecticut ordered a complete set of high-resolution aerial photographs. A second more detailed chemical and physical analysis of 70 lakes was undertaken in the 1970s (Norvell and Frink, 1975; Frink and Norvell, 1984). In addition, aerial flyovers were done in both 1970 and 1975. Recently, Canavan and Siver (1994) completed a third detailed study of the chemical and physical conditions of 60 Connecticut lakes. In addition, Field et al. (1996) used the aerial photographs from 1934, 1970, and 1990 to estimate the degree of change in the percentages of forest, agricultural/open field, and residential/urban lands in 30 of the watersheds.

Frink and Norvell (1984) compared their results with those of Deevey (1940) and concluded that Connecticut lakes were undergoing "advanced eutrophication." Frink and Norvell observed that total phosphorus concentra-

tions had increased in 33 of 35 lakes since the Deevey study, and that transparency had declined in most of the waterbodies. Frink and Norvell (1984) further observed that few changes in lake water alkalinity had occurred since the 1930s, and concluded that acidic deposition has had little impact on Connecticut lakes. Canavan and Siver (1994) compared lake water conditions, including trophic conditions and the composition of dissolved salts, between different geological regions.

The State of Connecticut is divided into five primary geological zones, the Eastern Uplands, Western Uplands, Coastal Slope, Central Valley and Marble Valley (see Rogers et al., 1959; Bell, 1985). The bedrock of the Eastern and Western Uplands and Coastal Slope is crystalline in nature and composed primarily of schists, gneiss and to a lesser extent granites (Rogers et al., 1959; Bell, 1985; Jokinen, 1983). The Marble and Central Valleys are composed mainly of metamorphosed limestone and brownstone, respectively (Rogers et al., 1959; Bell, 1985). In addition, exposed igneous basalt or trap rock sills are aligned in a north-south position within the Central Valley. Canavan and Siver (1994) found significant differences in the compositions and quantities of dissolved salts of lakes situated in the different geological regions. However, few differences in trophic variables were observed in lakes between the different geological regions (Canavan and Siver, 1994).

The purpose of this paper is to identify the degree and direction of change, if any, in the chemical and physical structure of Connecticut lakes over the last 55 yr through a comparison of the three surveys. Comparisons between the three data sets will include the variables Secchi disk depth, total phosphorus, and alkalinity. Comparisons in concentrations of base cations and chloride levels will also be made between the Frink and Norvell (1984) and Canavan and Siver (1994) studies. When possible, changes will also be related to shifts in land use in the surrounding watersheds as documented by Field et al. (1996).

MATERIALS AND METHODS

Data from 42 lakes examined by Canavan and Siver (1994) are included in this survey, 29 and 41 of which were common with the studies of Deevey (1940) and Frink and Norvell (1984), respectively. A total of 35 lakes were surveyed both by Deevey (1940) and Frink and Norvell (1984). Details on the location of each waterbody, methods employed, and all chemical and physical data used in this study can be found in Deevey (1940), Norvell and Frink (1975), Frink and Norvell (1984), and Canavan and Siver (1994; 1995). For simplicity, we refer to the Deevey (1940), Frink and Norvell (1984) and Canavan and Siver (1994) surveys as the 1930s, 1970s, and 1990s, respectively.

Direct comparisons of data between the three surveys were made for total phosphorus and Secchi disk depth measurements. In each of the three studies total phosphorus concentrations were determined colorimetrically by complexing with molybdenum after initial acid persulfate digestion of organic material.

Botany Department, Connecticut College, New London, CT 06320. Received 13 Feb. 1995. *Corresponding author (pasiv@conncoll.edu).

The methods for total phosphorus were essentially identical for the 1970s and 1990s, and in an earlier paper Norvell and Frink (1975) had determined that their method was comparable to that of Deevey. In each study a 20-cm Secchi disk was used, thus, no correction due to differences in size was necessary (Eilers et al., 1989).

Eilers et al. (1989) listed the two most common sources of variation in comparing alkalinity measurements to be due to sample storage and the method of titration employed. Kramer and Tessier (1982) determined that storage of samples in unrefrigerated soft glass bottles can result in an increase in alkalinity. Although it is unknown if Deevey used glass containers in the 1930s survey, he did process samples soon after collection, normally within a 1-d period (E.S. Deevey, Jr., 1996, personal communication). Polyethylene bottles were consistently used for the 1970s and 1990s surveys and samples were kept cold until analysis; in addition, polyseal caps were used in the 1990s. As a result, no corrections were felt necessary as a result of storage of unrefrigerated samples in glass containers.

Adjustments were necessary, however, due to differences in titration methods (see Kramer and Tessier, 1982; Kramer et al., 1986; Asbury et al., 1989; Eilers et al., 1989 for extensive discussions). The methyl orange method was used for alkalinity determinations in the 1930s and 1970s, making these data directly comparable (Norvell and Frink, 1975); such a comparison assumes that the same endpoint pH (color) was used in each case (Kramer et al., 1986). However, the Gran titration method, as described by Wetzel and Likens (1991), was used during the 1990s study. The pH at the equivalence point of the titration is lower as the alkalinity of the sample increases (Asbury et al., 1989; Eilers et al., 1989; Kramer et al., 1986). The Gran titration method accounts for differences in the pH at the equivalence point, whereas the methyl orange method assumes a fixed endpoint that in the case of dilute softwater lakes is below the pH at the equivalence point (Kramer et al., 1986). Thus, the methyl orange method results in an overestimation of alkalinity (Asbury et al., 1989). The degree of overestimation becomes less as the alkalinity of the sample increases. Thus, when comparing the 1990s data to the earlier two studies we had to decide the magnitude of the correction necessary, and which lakes needed to be corrected. We used the correction factor of $56 \mu\text{eq L}^{-1}$ proposed by Asbury et al. (1989) for softwater Adirondack lakes, and applied this correction to lakes with a methyl orange alkalinity of $<150 \mu\text{eq L}^{-1}$ (J.M. Eilers, personal communication). The correction of $56 \mu\text{eq L}^{-1}$ is similar to the $57 \mu\text{eq L}^{-1}$ correction made by Eilers et al. (1989) for northern Wisconsin lakes with a similar range in pH to those waterbodies examined in this study. We were unable to apply the correction method proposed by Kramer and Tessier (1982) due to the lack of necessary data. We recognize that using a correction factor of $56 \mu\text{eq L}^{-1}$ will influence the estimated change in alkalinity.

Direct comparisons were also made for concentrations of base cations and chloride between the 1970s and 1990s studies. Concentrations of base cations were determined on filtered and acidified samples in both surveys by comparable atomic absorption methods. Because concentrations of K^+ were very low compared to Na^+ , Ca^{2+} , and Mg^{2+} concentrations (Frink and Norvell, 1984; Canavan and Siver, 1994) and not found to be significantly different between the two studies, they will not be discussed further. Chloride levels were also estimated in both studies using the argentometric method with titration of the sample with silver nitrate to a standard color (APHA, 1971, 1985). Although data on chlorophyll concentrations were available for the 1930s, we made no comparisons because of

unresolvable differences in methods (see Norvell and Frink, 1975).

Changes in land use patterns referred to in this communication were determined from aerial photographs for 1934, 1970, and 1990 as outlined in Field et al. (1996). Land use was classified as water/wetland, forest, urban/residential and agricultural/open field. In practically all cases changes in the urban/residential land use class were the result of building of residential units. Detailed findings of the changes in land use since the 1930s are discussed in Field et al. (1996).

All analyses in this communication are based on samples taken only from surface waters. Collections from the 1930s and 1990s represent samples taken at 1 m (E.S. Deevey, Jr., 1996, personal communication; Canavan and Siver, 1994); those from the 1970s were taken from either 0.2 m or at several depths within the epilimnion or euphotic zone (Frink and Norvell, 1984). For comparison of total phosphorus and Secchi disk depths only summer data, usually between late June and early August, were used for the 1970s and 1990s surveys; this included approximately two or three collections for the 1970s and four for the 1990s. Secchi disk measurements for the 1930s study represent means of generally two or sometimes three values taken during the summer (E.S. Deevey, Jr., 1996, personal communication). Total phosphorus measurements from the 1930s represent mostly means of two or three collections taken during summer, however, the means for some of the lakes also include a single winter value (Deevey, 1940; E.S. Deevey, Jr., 1996, personal communication). Unfortunately, due to the reporting of means, and the lack of the original data, we were unable to remove the few winter readings.

Frink and Norvell (1984) reported means for alkalinity, base cations, and chloride that represented measurements made in summer, spring, and fall. For comparative purposes we pooled seasonal data for alkalinity and base cations for the 1990 data; pooling the 1990s data for alkalinity and base cations was reasonable because there was no significant difference between warm (summer) and cool months (spring and fall). Chloride measurements for the 1990s represent summer means. Canavan and Siver (1994) pooled data for 1991, 1992, and 1993 since there was no significant difference between years.

All ANOVA and regression and correlation analyses were done with SPSSX (Nörusis, 1985), and unless otherwise stated a 0.05 level of significance was used. Tests of significance for selected variables on individual lakes (i.e., between the 1930s and 1990s) were not possible because only mean values were available from the 1930s.

RESULTS

Secchi Disk Depth

There has been a distinct decline in the Secchi disk depth of Connecticut lakes between the 1930s and 1970s, and between the 1970s and the early 1990s (Tables 1 and 2; Fig. 1). The mean Secchi disk depth for the study lakes included in each survey has dropped from 4.9 m in the 1930s, to 4.3 m in the 1970s, to 3.6 m in the early 1990s (Table 1). Of the 29 lakes common between the Deevey study and the Frink and Norvell survey, 16 had decreased in Secchi disk depth, while four and eight remained unchanged or increased, respectively (Fig. 1A). On average, the Secchi disk depth decreased by 0.4 m between the 1930s and the 1970s.

The trend in declining Secchi disk depth observed

Table 1. The mean and range for Secchi disk depth, total phosphorus, alkalinity, base cation concentrations (total cations), sodium, calcium + magnesium, and chloride for the 1930s, 1970s, and 1990s. Values are computed using only the lakes that were common between at least two of the three time periods.

Parameter	1930s		1970s		1990s	
	Mean	Range	Mean	Range	Mean	Range
Secchi depth, m	4.9	1.1-10.0	4.3	1.8-8.2	3.6	0.9-7.6
Total phosphorus, † $\mu\text{g-P L}^{-1}$	12	4-31	15	5-35	24	9-46
‡	12	4-31	17	5-107	26	9-96
Alkalinity, meq L^{-1}	0.49	0.03-2.20	0.44	-0.02-2.19	0.52	0.00-2.41
Total cations, meq L^{-1}	-	-	0.96	0.37-2.64	1.03	0.20-3.1
Sodium, meq L^{-1}	-	-	0.27	0.12-0.95	0.32	0.10-1.07
Calcium + magnesium, meq L^{-1}	-	-	0.63	0.20-2.35	0.65	0.09-2.53
Chloride, meq L^{-1}	-	-	0.22	0.05-0.84	0.30	0.05-1.19

† Silver and Roseland removed from measurements.

‡ Includes Silver and Roseland.

between the 1930s and 1970s has become even more evident since the 1970s (Tables 1 and 2; Fig. 1). Twenty-eight of 41 waterbodies have declined in Secchi disk depth since the 1970s, while only five had improved light penetration of more than 0.5 m. Four of the five lakes with improved Secchi disk depths are dilute water-

bodies located in upland regions with watersheds that have remained heavily forested. On average, lakes have declined in Secchi disk depth by an additional 0.83 m since the 1970s.

Eighty-three percent of the lakes in the current study that were common with the Deevey survey have de-

Table 2. Differences in Secchi disk depths, total phosphorus concentrations, and alkalinity measurements between the 1990s and 1970s (90-70), and the 1990s and 1930s (90-30) for 42 Connecticut lakes. A positive or negative value denotes an increase or decrease, respectively, in that variable relative to the more recent time period.

Lake	Secchi depth, m		Total phosphorus, $\mu\text{g L}^{-1}$		Alkalinity, meq L^{-1}	
	90-70	90-30	90-70	90-30	90-70	90-30
Alexander	-1.4	-2.2	21	18	0.096	-0.028
Amos	-1.3		17		0.060	
Ball	0.1	-0.1	19	28	0.300	0.710
Bantam	-0.1	-0.6	7	24	0.000	0.038
Bashan	0.5	-2.2	7	-5	-0.014	-0.144
Beach	-2.6		6		0.036	
Beseck	-0.3	1.0	-2	1	0.060	0.046
Bigelow	0.1		6		0.026	
Billings	0.1		9		0.066	
Black (M)†	1.0	-0.7	10	6	0.170	0.211
Black (W)‡	1.2		10		0.026	
Candlewood	-2.6	-0.5	12	17	0.150	0.457
Crystal (E)§	-0.8	-0.8	7	8	0.056	-0.062
East Twin	-1.0	-2.0	16	21	0.260	0.246
Gardner	-1.3	-1.2	10	13	-0.060	-0.063
Green Falls		2.2		6		-0.160
Hayward	0.3	-1.8	7	11	-0.040	-0.103
Highland	-1.7	-1.2	5	1	0.070	0.010
Kenosia	0.2		11		0.990	
Linsley	-1.7	-0.8	-4	3	0.070	0.660
Long	-0.9	-0.1	-4	1	0.010	-0.070
Mashapaug	-3.2	1.5	8	9	0.046	-0.077
Mt. Tom	0.1	-0.9	15	14	0.030	0.015
Norwich	-0.5		6		-0.160	
Pachaug	-1.7		20		0.146	
Pataganset	-0.1	-1.3	5	6	-0.040	-0.070
Powers	0.8	0.0	6	7	-0.004	-0.186
Quassapaug	-2.9	-3.5	4	11	0.036	-0.080
Quonnipaug	-0.5	-6.1	0	8	-0.120	0.080
Rogers	-0.8	-2.0	8	13	-0.070	-0.101
Roseland	-1.9	-1.6	58	83	0.070	0.035
Silver	-0.4	0.3	-74	3	0.650	0.290
Squantz	-0.3		8		0.160	
Terramuggus	-0.8	-0.8	2	4	0.060	0.070
Tyler	-0.8		23		0.140	
Uncas	-1.6		11		-0.070	
Waramaug	-0.7	-2.5	22	37	0.030	0.100
Waumgumbaugh¶	-3.0	-3.0	23	22	0.030	0.306
West Hill	0.6	-0.5	5	1	0.066	-0.052
West Side	-0.5		27		0.020	
Wononscopomuc	-2.9	-3.9	7	13	0.220	0.214
Wyassup	-0.6		14		0.076	
Mean	-0.8	-1.2	9	13	0.090	0.080

† Located in the town of Meriden.

‡ Located in the town of Woodstock.

§ Located in the town of Ellington.

¶ Also known as Coventry.

creased in Secchi disk depth (Table 2; Fig. 1B). Twenty-one of 29 waterbodies have declined in Secchi disk depth since the 1930s by more than 0.5 m; all of these lakes have increased in total phosphorus concentration, on average by $16 \mu\text{g L}^{-1}$ (see below). Fourteen of the 21 lakes have also increased in alkalinity since the 1930s survey. Data on land use in the surrounding watersheds for 1934 and 1990 are available for 15 of the 21 waterbodies that experienced a decline in Secchi disk depth of more than 0.5 m (see Field et al., 1996). Even though the mean percentage of forest cover has only dropped 1.5% since 1934 in the 15 watersheds, the mean amount of residential/urban land has increased 15.5%, mostly at the expense of agricultural and/or old fields. Three lakes had improved light penetration of more than 1 m since the 1930s (Table 2). Despite the increased Secchi disk depths the total phosphorus levels had increased in each lake, but only by an average of $5 \mu\text{g L}^{-1}$. Two of the three lakes, Green Falls and Mashapaug, have declined in alkalinity since the 1930s (see below) and are situated in watersheds that have remained heavily forested.

Total Phosphorus Concentrations

Coupled with a decline in Secchi disk depth has been a significant increase in total phosphorus concentrations since the 1930s. Because the concentration of total phosphorus for Silver Lake in the 1970s ($107 \mu\text{g L}^{-1}$) and

Roseland Lake in the 1990s ($96 \mu\text{g L}^{-1}$) were about 4 standard deviation units above the mean, they were removed from the analysis. The mean concentrations of total phosphorus for the study lakes included in each survey have increased from $12 \mu\text{g L}^{-1}$ ($n = 29$) in the 1930s, to $15 \mu\text{g L}^{-1}$ ($n = 41$) in the 1970s, to $24 \mu\text{g L}^{-1}$ in the early 1990s ($n = 42$) (Table 1; Fig. 2). Between the 1930s and 1970s 19 of 26 lakes increased in total phosphorus by a mean of $6 \mu\text{g L}^{-1}$ (Fig. 2A). When compared to data from the 1990s, the total phosphorus concentrations have increased in all but one lake since the 1930s by an average of $11 \mu\text{g L}^{-1}$. Land use data are available for 9 of the 13 lakes that have experienced a $>10 \mu\text{g L}^{-1}$ increase; these watersheds have increased from a mean of 2% residential/urban lands to 18% since the 1930s. Lakes with the smallest increase in total phosphorus are primarily those that have remained mostly forested or experienced little change in land use.

Phosphorus levels have remained the same or declined in only four lakes between the 1970s and 1990s (Table 2; Fig. 2B). However, 51 and 22% of the lakes have increased between 1 and $10 \mu\text{g L}^{-1}$, and 11 and $20 \mu\text{g L}^{-1}$, respectively.

Alkalinity

Based on the lakes in common between the three surveys there was only a slight change in the mean

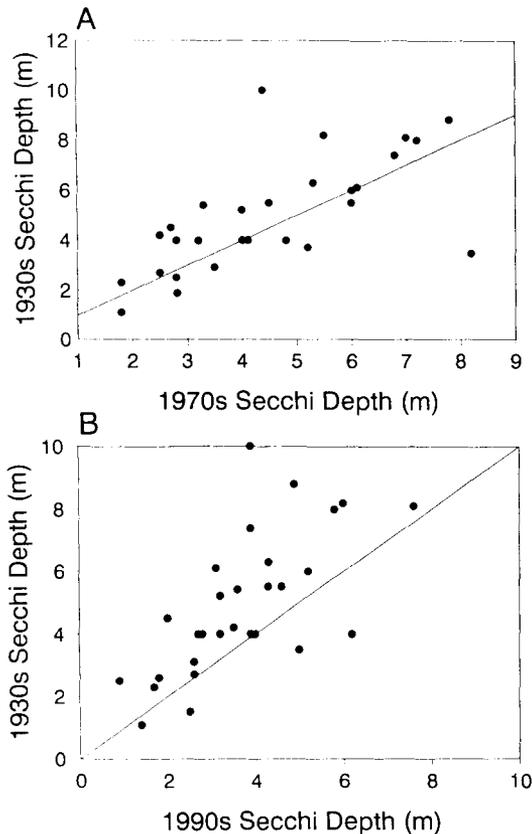


Fig. 1. Changes in the Secchi disk depths of Connecticut lakes between (A) the 1930s and 1970s, and (B) the 1930s and 1990s. The line represents a 1:1 ratio. Points positioned near the line indicate little or no change. Points lying above or below the line indicate that the value for that lake was greater in the 1930s when compared to the 1970s or 1990s, respectively.

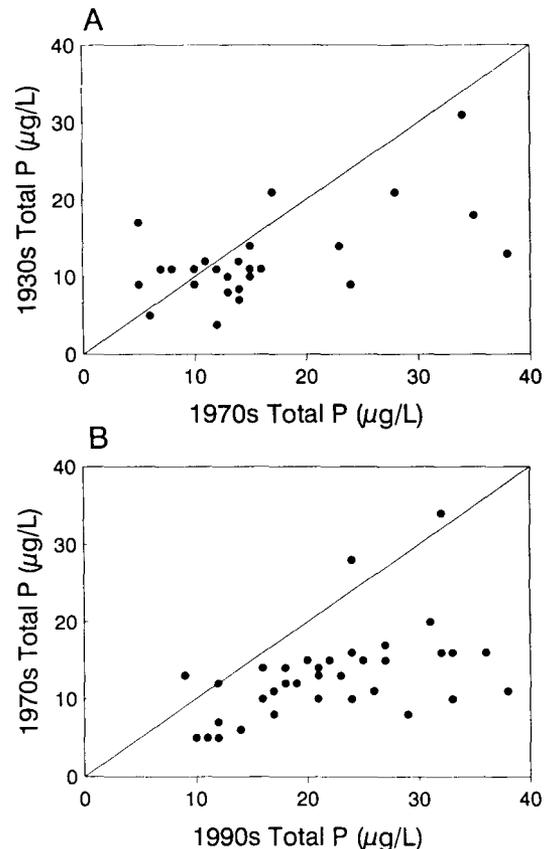


Fig. 2. Changes in the total phosphorus concentrations of Connecticut lakes between (A) the 1930s and 1970s, and (B) the 1970s and 1990s. The line represents a 1:1 ratio; see Fig. 1 for an explanation. Silver and Roseland were removed due to extraordinarily high levels.

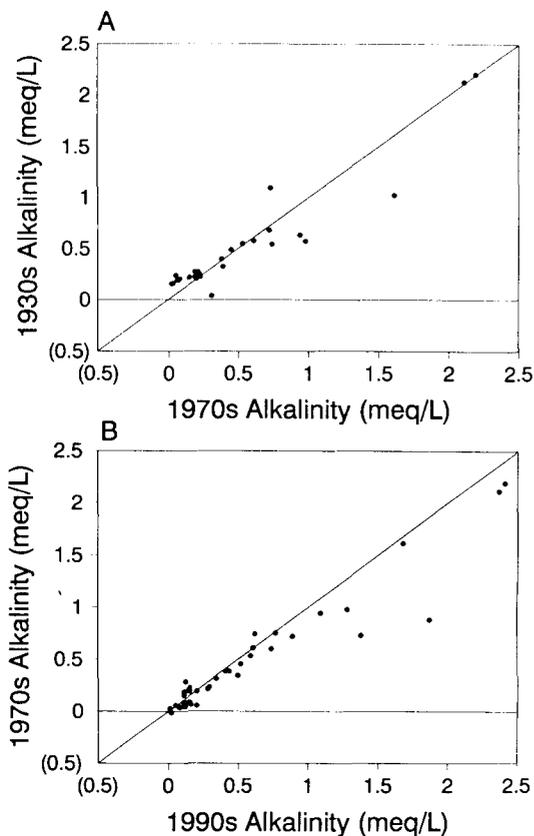


Fig. 3. Changes in the alkalinity of Connecticut lakes between (A) the 1930s and 1970s, and (B) the 1970s and 1990s. The line represents a 1:1 ratio; see Fig. 1 for an explanation. For the 1930s and 1970s data lakes with alkalinities below $150 \mu\text{eq L}^{-1}$ were corrected according to Asbury et al. (1989). See text for details.

alkalinity of Connecticut lakes since the 1930s (Tables 1 and 2; Fig. 3). The mean alkalinity for all study lakes included in each individual survey was 0.49, 0.44, and 0.52 meq L^{-1} for the 1930s, 1970s, and 1990s, respectively. There was a mean increase of 0.09 meq L^{-1} for lakes in common between the 1930s and 1990s studies. Of the 28 lakes common between each study seven and five showed a continuous increase or decline, respectively, in alkalinity, while nine waterbodies had little change. Curiously, seven lakes declined in alkalinity between the 1930s and 1970s, but then increased since about 1975.

Despite the similar mean alkalinity values for the three time periods, there was a group of lakes that experienced a significant decline in alkalinity, and a group that has significantly increased (Table 2). Thirteen of 29 lakes examined in both the 1930s and 1990s decreased in alkalinity between 5 and 100%, with a mean of 42% (Fig. 4). Each of the 13 lakes was situated in the Eastern or Western Uplands, or on the Coastal Slope, geological areas composed of highly resistant crystalline bedrock. The five lakes with the largest decline in alkalinity, Bashan, Green Falls, Hayward, Powers, and Rogers, are all situated in the eastern part of the state.

In contrast, only 9 of 41 lakes decreased in alkalinity between the 1970s and 1990s (Table 2; Fig. 3B). Of the 13 lakes that had declined in alkalinity between the 1930s

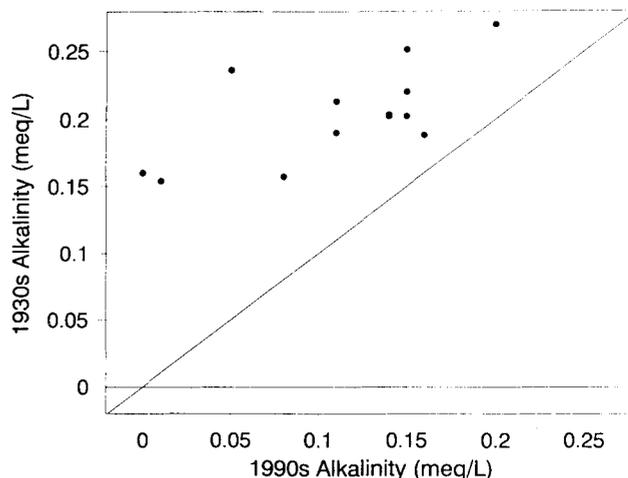


Fig. 4. Changes in alkalinity of a subgroup of low alkalinity lakes between the 1930s and 1990s. The line represents a 1:1 ratio; see Fig. 1 for an explanation. Alkalinity values below $150 \mu\text{eq L}^{-1}$ were corrected according to Asbury et al. (1989). See text for details.

and 1990s, only six (of 12 since Green Falls was not surveyed in the 1970s) actually declined since the 1970s (Table 2). That is, only six lakes had a unidirectional decline in alkalinity since the 1930s, whereas the others have increased since the 1970s. Bashan, Powers, Rogers, and Norwich experienced the largest percentage decrease in alkalinity since the 1970s.

A second set of 16 (of 29) waterbodies increased in alkalinity since the 1930s (Table 2; Fig. 3). Six of these lakes, Ball, Black-M, Candlewood, Linsley, Waramaug and Coventry, increased in alkalinity by more than 30% since the 1930s. Land use data are available for both 1934 and 1990 for 11 of the 16 watersheds (see Field et al., 1996). In all cases the percentage of residential land increased between 5 and 36%, with a mean increase of more than 20%. The four waterbodies with the greatest increase in alkalinity since the 1930s, Ball, Candlewood, Linsley, and Coventry, are all situated in highly developed watersheds and have had a mean increase in residential land of more than 30%. In addition, Lake Kenosia, another waterbody with a highly urbanized watershed, more than doubled in alkalinity since the 1970s.

Dissolved Ions

Total Cations. Lakes in common between the 1970s and 1990s surveys have experienced a mean increase in total cations of $70 \mu\text{eq L}^{-1}$; this represents a 9% rise (Tables 1 and 3; Fig. 5). Nine waterbodies have experienced a >20% increase in total cations (Table 3); each of these lakes have also had significant increases in residential land cover in their surrounding watersheds. Ten lakes, all located in either the Eastern Uplands or on the Coastal Slope, had a >20% decrease (Table 3; Fig. 5); these lakes have watersheds that have remained mostly forested (see below).

Sodium and Chloride. On average, the concentration of sodium in Connecticut lakes increased by $60 \mu\text{eq L}^{-1}$ since the 1970s, representing a rise of 23% (Table 3;

Table 3. Differences in concentrations of base cations (total), sodium, calcium + magnesium, and chloride between the 1970s and 1990s for 42 Connecticut lakes. A positive or negative value denotes an increase or decrease, respectively, in that variable since the 1970s. Absolute (Abs) and percent (%) changes are given. All values are in $\mu\text{eq L}^{-1}$.

Lake	Total		Na ⁺		Cl ⁻		Ca ²⁺ + Mg ²⁺	
	Abs	%	Abs	%	Abs	%	Abs	%
Alexander	-0.03	-8	-0.01	-8	-0.01	-17	-0.02	-10
Amos	-0.17	-15	0.06	22	0.06	23	-0.03	-5
Ball	0.80	43	0.67	168	0.45	61	0.11	8
Bantam	0.01	1	0.05	18	0.09	45	-0.04	-5
Bashan	-0.24	-39	-0.05	-23	0.04	33	-0.19	-50
Beach	-0.11	-25	-0.01	-4	0.01	6	-0.11	-52
Beseck	0.24	21	0.08	25	0.24	120	0.16	20
Bigelow	-0.33	-53	-0.10	-37	-0.05	-25	-0.22	-67
Billings	-0.15	-30	-0.04	-22	-0.02	-25	-0.11	-38
Black (M)†	0.37	24	0.26	67	0.03	5	0.12	11
Black (W)‡	-0.11	-20	0.00	0	0.14	88	-0.11	-37
Candlewood	0.28	19	0.09	32	0.21	105	0.19	17
Crystal (E)§	0.29	46	0.31	111	0.28	104	-0.03	-9
East Twin	0.07	3	-0.01	-8	0.01	12	0.08	4
Gardner	0.04	7	-0.01	-4	0.01	6	0.04	16
Green Falls	-	-	-	-	-	-	-	-
Hayward	0.07	17	0.04	21	0.11	183	0.03	15
Highland	0.17	21	0.10	29	0.14	35	0.08	18
Kenosia	1.55	100	0.73	251	0.77	285	0.80	66
Linsley	-0.01	<-1	-0.01	-1	0.11	39	-0.01	-1
Long	0.11	21	0.00	0	0.02	14	0.11	38
Mashapaug	-0.15	-23	0.01	4	-0.05	-14	-0.16	-46
Mt. Tom	-0.01	-1	0.00	0	0.03	9	-0.01	-2
Norwich	-0.28	-44	-0.08	-42	0.04	80	-0.21	-49
Pachaug	-0.04	-8	0.01	6	0.00	0	-0.06	-19
Pataganset	0.01	2	-0.03	-12	0.05	46	0.03	15
Powers	-0.27	-49	-0.16	-59	0.01	17	-0.11	-42
Quassapaug	-0.05	-10	-0.03	-17	-0.02	-12	-0.02	-7
Quonnipaug	-	-	-0.02	-9	0.07	50	-	-
Rogers	-	-	-	-	0.01	6	0.01	4
Roseland	-0.07	-8	0.01	4	0.13	76	-0.08	-13
Silver	0.44	22	0.34	71	0.13	16	0.10	7
Squantz	0.19	27	0.06	30	0.16	94	0.13	28
Terramuggus	0.03	3	0.02	4	0.34	97	0.00	0
Tyler	0.06	6	-0.04	-24	0.01	11	-0.11	15
Uncas	-0.19	-34	-0.09	-45	-0.01	-11	-0.11	-32
Waramaug	0.02	3	0.00	0	0.10	71	0.01	2
Waumgumbaugh¶	0.13	14	0.14	41	0.06	19	-0.01	-2
West Hill	0.02	5	0.01	8	0.06	100	0.05	23
West Side	-0.25	-19	-0.01	-6	0.08	89	-0.24	-21
Wononscopomuc	0.22	8	0.04	16	0.03	13	0.18	8
Wyassup	-0.12	-24	-0.02	-10	0.02	22	-0.11	-38
Mean	0.07	9	0.06	15	0.10	47	0.01	-6

† Located in the town of Meriden.

‡ Located in the town of Woodstock.

§ Located in the town of Ellington.

¶ Also known as Coventry.

Fig. 6A). Fourteen of 40 lakes have increased in sodium by more than 16%; six of these waterbodies, Kenosia, Ball, Crystal-E, Silver, Black-M, and Coventry, have increased between 41 and 251%, with a mean of 410 $\mu\text{eq L}^{-1}$ (Table 3).

Despite the overall rise in sodium levels, 10 lakes had decreased concentrations of more than 10% (Table 3). Four lakes in particular with undeveloped watersheds, Bigelow, Norwich, Powers, and Uncas, have sodium levels that are more than 37% lower than in the 1970s; this represents a mean decline of 110 $\mu\text{eq L}^{-1}$.

A total of 34 of 41 lakes had an increase in chloride levels since the 1970s, representing a mean increase of 90 $\mu\text{eq L}^{-1}$ (Tables 1 and 3; Fig. 6B). Fifteen localities increased by 50% or more, and only six lakes had a decrease. Six localities, Ball, Beseck, Candlewood, Crystal-E, Kenosia, and Terramuggus, had between a 210 and 770 $\mu\text{eq L}^{-1}$ increase; each of these lakes has a high degree of developed watershed.

Strong relationships were observed between the concentrations of sodium and chloride for the 1970 and 1990 periods. For measurements from the 1990s the correlation between sodium and chloride concentrations had a slope close to unity (0.99) and an r^2 of 0.84 (Fig. 7A). If one lake, Linsley Pond, with a known source of sodium from a trap rock quarry is removed, the relationship has a slope of 1.1, and a significantly higher r^2 of 0.96. Likewise, the removal of Linsley Pond from the 1970 data set yielded a significant relationship with an r^2 of 0.7, but a slope significantly greater than one (1.77). There was also a significant relationship between the change in sodium content in a lake since the 1970s and the change in chloride over the same period (Fig. 7B); this relationship had a slope of 1.1 and an r^2 of 0.67.

Calcium and Magnesium. Even though the mean concentration of Ca and Mg has remained relatively constant since the 1970s (Table 1), a group of lakes have

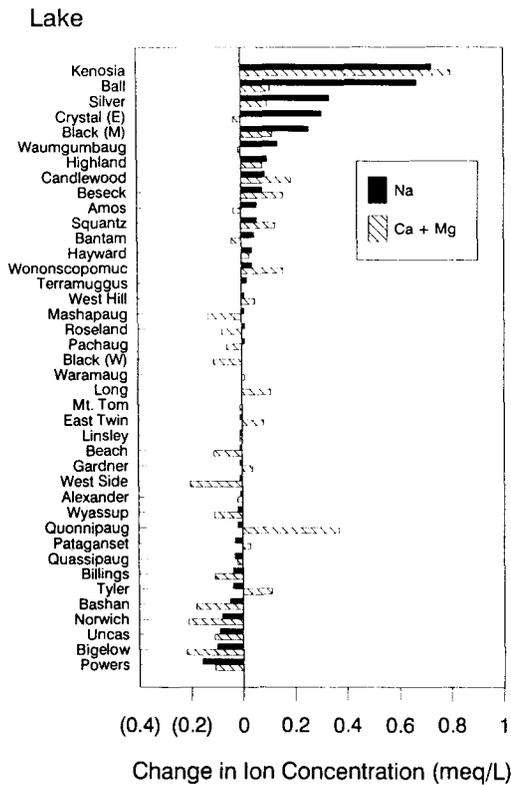


Fig. 5. Changes in the concentrations of sodium and divalent ions (calcium and magnesium) of 40 Connecticut lakes between the 1970s and 1990s. Lakes are arranged according to changes in sodium levels.

had a significant increase and another set a significant decrease (Table 3; Fig. 5). A total of 21 and 18 lakes have decreased or increased, respectively, in concentration since the 1970s. Ten waterbodies, all situated in the Eastern Uplands or on the coastal slope, have had more than a 30% decline in Ca and Mg levels, representing a mean decrease of $144 \mu\text{eq L}^{-1}$. Five lakes had a >20% increase, ranging between 50 and $800 \mu\text{eq L}^{-1}$.

DISCUSSION

It is evident that there has been an overall increase in the trophic state of Connecticut waterbodies since the late 1930s. Because Canavan and Siver (1994) noted significant correlations in Connecticut lakes between the four trophic related variables, Secchi disk depth, total phosphorus, total nitrogen and chlorophyll-*a* concentrations, we believe the historical trends in Secchi disk depth and total phosphorus can be valuable tools in tracking changes in trophic state. Also, since the percentages of wetlands in the watersheds and the general morphological features of the waterbodies have not significantly changed between the 1930s and 1990s (Field et al., 1996), we believe that changes in water color have played a minor role in controlling the observed declines in Secchi disk depth.

On average, lakes have lost more than 1.2 m in Secchi disk depth and have more than doubled in their concentrations of total phosphorus. A greater percentage of the decline in Secchi disk depth occurred since the 1970s

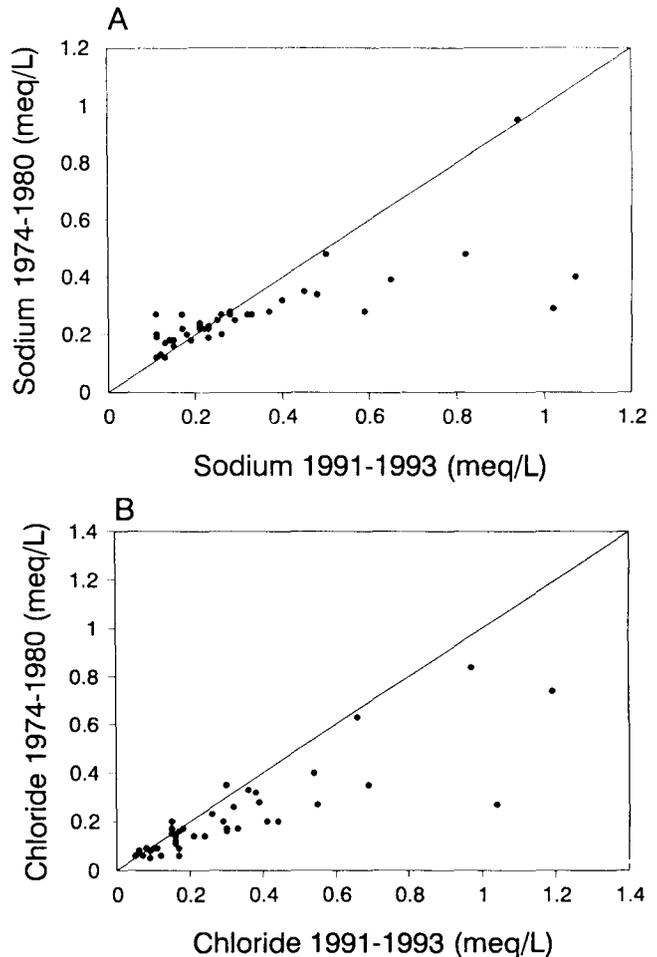


Fig. 6. Changes in (A) sodium and (B) chloride concentrations for Connecticut lakes between the 1970s and 1990s. The line represents a 1:1 ratio; see Fig. 1 for an explanation.

(-0.8 m) than between the 1930s and 1970s (-0.4 m). When Deevey made his survey in the 1930s 45% of the lakes had a Secchi disk depth more than 5 m, indicative of an oligotrophic or early mesotrophic condition (Frink and Norvell, 1984). In comparison, <10% of the 42 waterbodies in this survey had mean Secchi disk depths >5 m. Interestingly, the six lakes that Deevey noted to have Secchi disk depths <3 m (mean = 2.1 m) have similar Secchi disk depths today (mean = 1.9 m). We conclude that the trophic state of the more oligotrophic lakes with high light penetration have degraded the most since the 1930s.

The decline in Secchi disk depth since the 1930s has been unidirectional in 26 of the lakes, suggesting a continuous degradation of the trophic state. Likewise, the increases in the total phosphorus concentrations have also been unidirectional in the majority of lakes since the 1930s, and to a greater degree between the 1970s and 1990s. Using the trophic classification system of Frink and Norvell (1984), Connecticut lakes on the average have shifted from being oligotrophic-mesotrophic (1930s), to early mesotrophic (1970s), to late mesotrophic (1990s).

The increases in total phosphorus concentrations in

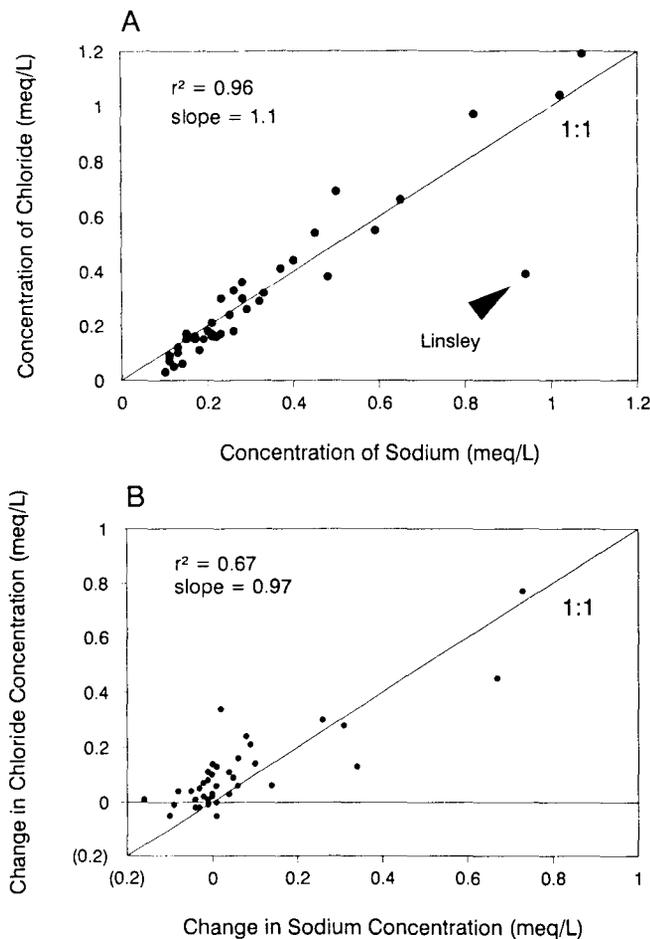


Fig. 7. (A) Correlation between sodium and chloride concentrations of Connecticut lakes in the 1990s. (B) Relationship between the changes in sodium and chloride concentrations of Connecticut lakes between the 1970s and 1990s.

Connecticut waterbodies since the 1930s are consistent with concentrations estimated using changes in land use patterns in the surrounding watersheds (Field et al., 1996). It is well documented that the export of phosphorus, and nitrogen, is greater from both residential/urban and agricultural lands than from forests (see Omernik, 1976; Wanielista et al., 1977; Hill, 1981; Beaulac and Reckhow, 1982; Osborne and Wiley, 1988; Frink, 1991 and references therein). Although widely different export coefficients for different land uses have been reported for phosphorus (Beaulac and Reckhow, 1982; Frink, 1991), they are often 5 to more than 10 times greater for urban than forested land (Frink, 1991).

In an analysis that included many of the lakes in our study Norvell et al. (1979) reported phosphorus export coefficients of 1.7, 0.54, and 0.1 kg phosphorus/ha per year for urban, agricultural, and forested lands, respectively. Norvell et al. (1979) further proposed a model relating the proportions of each land use type in a watershed to the concentration of total phosphorus in the receiving lake. Field et al. (1996) found that the mean percentage of residential/urban land in 30 of the watersheds changed from 2% to 10% to 16%, from 1934, to 1970 to 1990, respectively. Likewise, the change in the

mean percentage of agricultural land for the same three periods was 20, 11, and 7%, respectively. The percentage of forests was similar for the 3 yr. Applying the land use values for the three time periods to the Norvell et al. (1979) model yielded estimated mean total phosphorus levels of 15, 19, and 25 μg phosphorus L^{-1} for the 30 lakes (Field et al., 1996). The estimated concentrations of phosphorus are similar to the actual changes reported in this study for the three time periods, supporting the idea that the accelerated eutrophication of Connecticut lakes is indeed correlated with changes in land use in the surrounding watersheds.

Trends in alkalinity changes in Connecticut lakes over the last 55 yr are not as clear as trophic parameters. Although there was no significant difference between the mean alkalinity levels for lakes in common between the three surveys, there was a group of lakes where the concentrations had declined, and another where the concentrations had increased. Declines in the alkalinities of lakes in the northeastern USA, especially those adversely affected by acidic deposition, are well documented (see Charles, 1991 and references therein). Estimates of the amount of decline in lake water alkalinity levels and/or pH over the past ca. 100 to 300 yr have also been documented using paleolimnological inference methods for regions in the northeastern USA, including northern New England (Davis et al., 1990, 1994; Kahl et al., 1991) and the Adirondacks (Smol et al., 1984; Christie and Smol, 1986; Charles et al., 1987; Cumming et al., 1992). Dilute lakes in uninhabited catchments studied in northern New England were found to have increased in inferred pH and alkalinity during periods of logging within the catchments (ca. early 1800s to early 1900s); all of the lakes have declined in pH and alkalinity to below pre-logging levels since about 1915, presumably due to acidic deposition (Davis et al., 1994). Other lakes in Maine have showed little or no acidification (Kahl et al., 1991). Cumming et al. (1992) estimated that 80% of low alkalinity lakes in the Adirondacks have undergone decreases in pH and alkalinity since pre-industrial times.

Eilers and Selle (1991) reported the mean pH of precipitation in the Adirondacks, ME and the Catskills as 4.37, 4.5, and 4.28, respectively. According to Husar et al. (1991) and Sisterson (1991) much of the northeastern USA, including Connecticut, has received precipitation with a pH ranging from 4.2 to 4.6 over the last 10 yr. Despite the low pH of the precipitation and large areas composed of crystalline bedrock (Bell, 1985; Rogers et al., 1959), scientists have not observed significant declines in lakewater pH in Connecticut (CAES, 1983; Frink and Norvell, 1984). In addition, in a paleolimnological study of five Connecticut lakes situated in crystalline bedrock regions of the state, Marsicano and Siver (1993) did not detect significant declines in pH. Four of the five lakes studied by Marsicano and Siver (1993) showed only slight and nonsignificant changes in alkalinity since the 1930s; data were lacking for the fifth lake.

Although many lakes in Connecticut have not experienced declines in pH or alkalinity, comparisons made in this study indicate that a small population of lakes has declined in alkalinity since the 1930s. The group

of lakes includes five dilute waterbodies with current alkalinities between 0 and 0.15 meq L^{-1} , Green Falls, Bashan, Hayward, Powers, and Rogers, that have each continued to decrease in alkalinity by more than 40% since the 1930s. In addition, Norwich Pond has decreased in alkalinity by 57% since the 1970s. This group of Connecticut lakes share many characteristics with lakes in the northeastern USA with declining alkalinity histories. First, in the Adirondacks (Driscoll et al., 1991) and Maine (Kahl et al., 1991) many of the clearwater drainage lakes adversely affected by acidic deposition were ones situated in forested watersheds, with highly resistant granitic bedrock and thin layers of glacial till. Second, lakes often affected by acidic deposition are ones with relatively low watershed to lake surface area ratios that receive a relatively large percentage of their water load directly from precipitation (Driscoll et al., 1991; Kahl et al., 1991; Battarbee et al., 1988). Third, the development of humic rich acidic forest soils can further increase the acidification and decrease the alkalinity of runoff (Rosenqvist, 1978; Krug and Frink, 1983; Richter, 1984; Kramer et al., 1986). Each of the six dilute Connecticut waterbodies with unidirectional declining alkalinities is situated in a primarily forested watershed in a crystalline bedrock region. In all cases, the surrounding forests have continued to mature over the last ca. 50 to 60 yr. Five of the lakes also have a relatively small watershed to lake area ratio (Canavan and Siver, 1994).

Brakke et al. (1988) suggested that long-term acidification of lakes in the northeastern USA was related to increased sulfate concentrations. Increased concentrations of sulfate in surface runoff must be balanced with an increase in cation concentrations. In acidic forest soils where exchange sites are often dominated by H^+ ions (and to a lesser degree Al^{3+}) and mineral weathering is low, increased sulfate concentrations are generally balanced by H^+ , resulting in a decrease in alkalinity and an increase in acidity of surface runoff (Kramer et al., 1986). Since the watersheds of the group of lakes with declining alkalinities have remained primarily forested since the 1930s (Field et al., 1996), it is reasonable to assume that this mechanism is important in low alkalinity Connecticut lakes. Further study will be needed to determine why the concentrations of divalent base cations have decreased in many of the lakes with heavily forested watersheds and declining alkalinities. It is possible that supplies of these base cations have been leached from the acid soils over the past several decades, a mechanism discussed in detail by Kramer et al. (1986).

Many of the study lakes have significantly increased in alkalinity since the 1930s, despite receiving acidic deposition. We believe that the increases in alkalinity are largely the direct result of anthropogenic disturbances. Disturbances in the surrounding watersheds, such as the building of roads and homes, results in the removal of low pH, organic rich forest soils, and subsequent exposure of new base cation rich subsoils (Haines and Akielaszek, 1983; Eilers et al., 1989; Prowse, 1987). Thus, more of the strong acid anions in surface waters would be balanced with base cations instead of H^+ or Al^{3+} ions, resulting in an increase in alkalinity. Increased amounts

of chemical weathering resulting from slight decreases in soil pH as an indirect result of acid deposition can also result in increased alkalinity (Kilham, 1982). Increased residential development also results in increases in the amounts of lime used to support grass yards and gardens, in the application of fertilizers, and in wastewater runoff, all activities that can increase the alkalinity of surface runoff. Modern agricultural activities can also result in elevated alkalinity of runoff (Pollman and Canfield, 1991).

All of the lakes that have experienced large increases in alkalinity since the 1930s have all had significant increases in residential areas within their watersheds (see Field et al., 1996), supporting the hypothesis that urbanization of the watershed can result in an increase in the alkalinity of receiving waterbodies. This hypothesis has been advanced and supported by previous studies. Stoddard and Murdoch (1991) noted that long-term records indicated that alkalinity levels have increased in some streams in the Catskill (New York) Mountains, presumably as a result of land use changes. Renberg et al. (1993) provided paleolimnological evidence that human-related alterations in a watershed caused alkalization of surface waters in Sweden. Eilers et al. (1989) suggested that alkalinity increases in a set of Wisconsin lakes were directly related to watershed disturbances. Cumming et al. (1992) noted that diatom inferred alkalinities had increased in several Adirondack lakes, possibly related to human activity in the watersheds. In their study of five Connecticut lakes Marsicano and Siver (1993) concluded that increased urbanization had masked potential effects of acidic deposition. Davis et al. (1994) observed that logging resulted in increased buffering capacities of northern New England lakes. Thus, decreasing alkalinities observed in lakes in other parts of northeastern North America caused by acidic deposition may not be as evident in Connecticut because of higher degrees of urbanization.

Interactions between the cycling of iron, sulfur, and phosphorus may also represent potential mechanisms for increasing alkalinities and total phosphorus concentrations in Connecticut lakes, especially during summer stratification when the hypolimnia of many of the lakes become anaerobic (Canavan and Siver, 1995). Alkalinity can be generated through the bacterial mediated reduction of sulfate under anaerobic conditions (Kilham, 1982), a process known to occur in the anaerobic hypolimnia of some New England (Giblin et al., 1990) and Connecticut (Murray, 1994) lakes. If the sulfide is balanced by ferrous iron, a redox reactive cation, the increased levels of alkalinity are maintained provided that the hypolimnion remains anaerobic. Alkalinity generation caused by sulfate reduction would be further fueled through acidic deposition of sulfate.

Another indirect consequence of the binding and precipitation of iron cations with reduced sulfur is a reduction in the pool of iron cations available to precipitate phosphates from the water column. Thus, loading of sulfate by acidic deposition may also result in increased internal loading of phosphorus into the water column (Kilham, 1982; Murray, 1994). Further study would be

needed to determine the relative degrees to which changes in land use, sulfate loading, and other mechanisms such as increased chemical weathering (Kilham, 1982), have played roles in controlling changes in Connecticut lakes.

The majority of the mean increase in total cations in Connecticut lakes since the 1970s has been primarily the result of an increase in sodium concentrations, and to a lesser degree of increased levels of divalent cations. The increase in base cations has been balanced primarily by an increase in chloride, alkalinity ions, or both. In some lakes, especially low alkalinity lakes in the upland regions and along the coastal slope (Canavan and Siver, 1994), changes in sulfate concentrations may have been of importance.

Despite these overall trends, lakes differed widely on an individual basis. For example, the two lakes with the largest increase in total cations since the 1970s, Kenosia (+1550 $\mu\text{eq L}^{-1}$) and Ball (+800 $\mu\text{eq L}^{-1}$), experienced different patterns. In the case of Kenosia, increases in sodium (730 $\mu\text{eq L}^{-1}$) and divalent cations (800 $\mu\text{eq L}^{-1}$) were relatively balanced with increases in chloride (770 $\mu\text{eq L}^{-1}$) and alkalinity (990 $\mu\text{eq L}^{-1}$). However, in the case of Ball even though the increase in chloride and alkalinity was within 4% of the increase in base cations, there was considerably more sodium than chloride, suggesting that sodium cations may balance some of the increase in alkalinity ions.

Even though the mean increase in the concentration of sodium was 60 $\mu\text{eq L}^{-1}$, six waterbodies increased by an average of over 410 $\mu\text{eq L}^{-1}$. Five of the six lakes had an increase in the percentage of residential/urban land of between 4 and 16% since the 1970s; the sixth lake had little change in the percentage of residential/urban land since the 1970s, however, was already highly urbanized by the 1970s. Our results indicate a significant relationship between the increase in residential land in the watershed and the increase in lake sodium concentrations.

The question arises as to what processes would cause an increase or decrease in the concentrations of sodium and chloride in lake water. Sources of sodium and/or chloride include weathering of mineral substrates (e.g., see Likens et al., 1977), sea spray and precipitation (Sullivan et al., 1988), agricultural inputs (Dickinson, 1983), animal wastes (Wagner et al., 1976; Pionke and Urban, 1984), industrial inputs (Weibel et al., 1964; Prowse, 1987), and road deicing salts (Prowse, 1987; Mattson and Godfrey, 1994). Likens et al. (1977) estimated the annual input of sodium through weathering processes from an undisturbed forest to be 5.6 kg Na/ha. Sullivan et al. (1988) reported that many New England lakes had a Na/Cl ratio greater than that found in seawater, in part, due to weathering reactions in the watersheds. Prowse (1987) found that urbanization increased the concentrations of sodium and chloride above the "natural denudation rate" by as much as 104 and 63 times, respectively, most likely due to consumer waste, domestic waste and road salt sources. In a recent study, Mattson and Godfrey (1994) concluded that the use of NaCl as a road deicing salt accounted for the bulk of sodium (and chloride) in Massachusetts waterways. Mattson and Godfrey (1994) further estimated that sea spray/

precipitation and weathering processes accounted for only about 4 and 10% of the sodium concentrations in Massachusetts streams.

As was concluded in the Massachusetts study (Mattson and Godfrey, 1994), we believe that the primary source of both sodium and chloride in Connecticut lakes, and the source of the increased concentrations since the 1970s, is from road deicing salts. Several lines of evidence support this idea. First, for most lakes the ratio of concentrations of sodium and chloride ions (0.9:1) is close to the seasalt ratio in equivalents (0.86); such a relationship is a strong indication of road salt contamination (Mattson and Godfrey, 1994; Stoddard and Murdoch, 1991). Second, the correlation between the change in sodium levels since the 1970s and the change in chloride concentrations over the same time period is significant ($r^2 = 0.67$), and has a slope of 0.97. This indicates that for this suite of lakes an increase or decrease in sodium ions was often balanced by an equal increase or decrease in chloride ions. Third, the fact that significant relationships were observed between sodium or chloride concentrations in Connecticut lakes and the percentage of residential/urban development in the surrounding watersheds (see Field et al., 1996) is also a strong indication that urbanization was the primary cause of the increased sodium and chloride concentrations. An increase in residential development would undoubtedly result in an increase in the total linear distance of roads, and hence the use of road deicing salts. The amount of impervious surface that may also require use of deicing salts is further increased through the building of driveways and walkways. Since the water chemistry of lakes in Connecticut is similar to that for lakes in Massachusetts situated in similar geological settings (Canavan and Siver, 1994), it is reasonable to conclude that road salts may also be the primary input of sodium in Connecticut lakes.

Other sources of dissolved ions, including sodium and chloride, may be enhanced with increased urbanization. For example, further development of a watershed would inevitably result in increased weathering and subsequent leaching of newly exposed mineral substrates (Haines and Aklielaszek, 1983; Prowse, 1987; Sullivan, 1991). The increased leaching generally results in an elevation of base cation concentrations in surface waters (Norvell, 1977; Frink and Norvell, 1984; Eilers et al., 1989; Sullivan, 1991). Another potential source of dissolved ions to lakes is through contamination by faulty septic systems. Such a source is of increasing importance in this suite of lakes since many of the homes along the shorelines of the lakes are older, were originally built as seasonal units, and have inadequate septic designs (i.e., cesspools). Liming soils to improve acidity is a common practice in New England and provides an additional source of base cations, especially Ca, in waterways.

Another source of sodium, but not chloride, in lakes situated in the Central Valley is through weathering of basaltic trap rock (Norvell, 1977; Canavan and Siver, 1994). Unusually high levels of sodium and elevated Na/Cl ratios were reported for two lakes, Linsley and Cedar Ponds, during the 1970s (Norvell, 1977; Norvell

and Frink, 1975). The elevated sodium levels relative to chloride concentrations were the result of drainage from a trap rock quarry, which is still active today. In the current study, the Na/Cl ratio of Linsley Pond was widely divergent from all other lakes, but similar to that reported by Norvell (1977).

Despite the overall increase in sodium levels, a small number of waterbodies have experienced a decline. In general, the waterbodies with the greatest decreases in sodium concentrations are all situated in primarily wooded watersheds located in the Eastern Uplands or along the Coastal Slope. The four lakes with the largest decline in sodium, Bigelow, Uncas, Norwich, and Powers, are situated in primarily forested watersheds and have had less than a 1% increase in residential/urban land since 1970.

There are at least two potential reasons that may explain why sodium concentrations have declined in these few Eastern Upland lakes. First, many of the surrounding forests are relatively young, having developed from previously agricultural areas commencing near the turn of the century (Bell, 1985). It is possible that over the last 20 yr the continued maturation of the forest has sequestered base cations from the soils, and that the soils have become leached of a portion of their base cation supply (Likens et al., 1977; Nilsson et al., 1982). The second, and more likely reason, may be related to a change in the application procedure of deicing salts on roads since the 1970s. In general, since the 1970s there has been a shift towards using a mix of NaCl and sand for deicing purposes, especially in state forest lands where these lakes are situated. Thus, if the linear distance of roads were to remain constant, but the proportions of NaCl used in deicing procedures were to decrease, the subsequent runoff may have lower concentrations of salt.

Despite the changes observed in lake water chemistry in this study several other factors need to be considered. First, the estimated changes are based on relatively few data from the 1930s, making it difficult to thoroughly evaluate variability due to methodology and short-term (day to month) fluctuations. Second, the effect of differences in climate between the three time periods on the results is unknown. Any significant differences in the amounts and patterns of annual precipitation and temperature would undoubtedly influence the results, and in a differential manner because of differences in morphological features and retention times (Canavan and Siver, 1994).

In conclusion, the chemistry of many Connecticut lakes has significantly changed since the 1930s, especially for waterbodies situated in watersheds that have decreased in forest cover and become more urbanized. In general, these lakes have become less transparent, have increased in total phosphorus and dissolved salt content, and have maintained or increased in alkalinity. We believe that most of the increases in sodium and chloride concentrations were caused by use of road deicing salt. Anthropogenic related changes in the surrounding watersheds have most likely played a role in the increased

total phosphorus concentrations, decreased Secchi disk depths, and the alkalization of many of the study lakes.

ACKNOWLEDGMENTS

The late Professor Edward Deevey Jr. was very helpful in the early planning stages prior to funding of this project, and provided insights from his earlier experiences with Connecticut lakes. We thank J. Hartranft, C. Weillhoefer, and D. Bender for field and laboratory assistance. We also thank Joe Eilers, Tom Murray, Clyde Asbury, and Peter Rich for discussions on chemical issues, and several reviewers for excellent suggestions. This project was funded by the National Science Foundation (grant no. DEB-9306587), the Connecticut Department of Environmental Protection and the Institute of Water Resources.

REFERENCES

- American Public Health Association. 1971. Standard methods for the examination of water and wastewater. 13th ed. American Public Health Assoc., Washington, DC.
- American Public Health Association. 1985. Standard methods for the examination of water and wastewater. 20th ed. American Public Health Assoc., Washington, DC.
- Asbury, C.E., F.A. Vertucci, M.D. Mattson, and G.E. Likens. 1989. Acidification of Adirondack lakes. *Environ. Sci. Technol.* 23:362-365.
- Battarbee, R.W., N.J. Anderson, P.G. Appleby, R.J. Flower, S.C. Fritz, E.Y. Haworth, S. Higgitt, V.J. Jones, A. Kreiser, M.A.R. Munro, J. Natkanski, F. Oldfield, S.T. Patrick, N.G. Richardson, B. Rippey, and A.C. Stevenson. 1988. Lake acidification in the United Kingdom 1800-1986. *Ensis Publ.*, London.
- Beaulac, M.N., and K.H. Reckhow. 1982. An examination of land use-nutrient export relationships. *Water Resour. Bull.* 18:1013-1024.
- Bell, M. 1985. The face of Connecticut: People, geology and the land. State Geological and Natural History Survey of Connecticut Bull. 110.
- Brakke, D.F., D.H. Landers, and J.M. Eilers, 1988. Chemical and physical characteristics of lakes in the Northeastern United States. *Environ. Sci. Technol.* 22:155-163.
- Brenner, M., T.J. Whitmore, M.S. Flannery, and M.W. Binford. 1993. Paleolimnological methods for defining target conditions in lake restoration: Florida case studies. *Lake Reserv. Manage.* 7: 209-217.
- Canavan IV, R.W., and P.A. Siver. 1994. Chemical and physical properties of Connecticut lakes with emphasis on regional geology. *Lake Reserv. Manage.* 10:175-188.
- Canavan IV, R.W., and P.A. Siver. 1995. Connecticut Lakes: A study of the chemical and physical properties of fifty-six Connecticut Lakes. Connecticut College Arboretum, New London.
- Charles, D.F. ed. 1991. Acidic deposition and aquatic ecosystems: Regional case studies. Springer-Verlag New York, New York.
- Christie, C.E., and J.P. Smol. 1986. Recent and long-term acidification of Upper Wallface Pond (N.Y.) as indicated by mallowonadacean microfossils. *Paleolimnology IV: Proceedings of the Fourth International Symposium on Paleolimnology. Hydrobiologia* 143: 355-360.
- Charles, D.F., D.R. Whitehead, D.R. Engstrom, B.D. Fry, R.A. Hites, S.A. Norton, J.S. Owen, L.A. Roll, S.C. Schindler, J.P. Smol, A.J. Uutala, J.R. White, and R.J. Wise. 1987. Paleolimnological evidence for recent acidification of Big Moose Lake, Adirondack Mountains, N.Y. (U.S.A.). *Biogeochemistry* 3:267-296.
- Connecticut Agriculture Experiment Station. 1983. Acid rain: Sources and effects in Connecticut. Report of the Acid Rain Task Force to the Governor and to the General Assembly. Bull. 809.
- Cumming, B.F., J.P. Smol, J.C. Kingston, D.F. Charles, H.J. B. Birks, K.E. Camburn, S.S. Dixit, A.J. Uutala, and A.R. Selle. 1992. How much acidification has occurred in Adirondack region lakes (New York, USA) since preindustrial times? *Can. J. Fish Aquat. Sci.* 49:128-141.
- Davis, R.B., D.S. Anderson, M.C. Whiting, J.P. Smol, and S.S. Dixit. 1990. Alkalinity and pH of three lakes in northern New

- England, U.S.A. over the past 300 years. *Phil. Trans. R. Soc. London B* 327:413-421.
- Davis, R.B., D.S. Anderson, S.A. Norton, and M.C. Whiting. 1994. Acidity of twelve northern New England (U.S.A.) lakes in recent centuries. *J. Paleolimnol.* 12:103-154.
- Deevey Jr., E.S. 1940. Limnological studies in Connecticut. V. A Contribution to Regional Limnology. *Am. J. Sci.* 238:717-741.
- Dickinson, W.E. 1983. Salt products in North America and outlook for the future. p. 657-659. *In* B.C. Schreiber and H.L. Harner (ed.) Sixth International Symposium on Salt. Vol. II. Salt Inst., Alexandria, VA.
- Driscoll, C.T., R.M. Newton, C.P. Gubala, J.P. Baker, and S.W. Christensen. 1991. Adirondack Mountains. p.133-202. *In* D.F. Charles (ed.) Acidic deposition and aquatic ecosystems: Regional case studies. Springer-Verlag New York, New York.
- Eilers, J.M., and A.R. Selle. 1991. Geographic overview of the regional case study areas. p. 133-202. *In* D.F. Charles (ed.) Acidic deposition and aquatic ecosystems: Regional case studies. Springer-Verlag New York, New York.
- Eilers, J.M., G.E. Glass, A.K. Pollack, and J.A. Sorensen. 1989. Changes in conductivity, alkalinity, calcium, and pH during a 50-year period in selected northern Wisconsin lakes. *Can. J. Fish. Aquat. Sci.* 46:1929-1944.
- Field, C.K., P.A. Siver, and A.M. Lott. 1996. Estimating the effects of changing land use patterns on Connecticut lakes. *J. Environ. Qual.* 25:325-333 (this issue).
- Frink, C.R. 1991. Estimating nutrient exports to estuaries. *J. Environ. Qual.* 20:717-724.
- Frink, C.R., and W.A. Norvell. 1984. Chemical and physical properties of Connecticut Lakes. *Connecticut Agric. Exp. Stn. Bull.* 817.
- Giblin, A.E., G.E. Likens, D. White, and R.W. Howarth. 1990. Sulfur storage and alkalinity generation in New England lake sediments. *Limnol. Oceanogr.* 35:852-869.
- Haines, T.A., and J.J. Akielaszek. 1983. A regional survey of the chemistry of headwater lakes and streams in New England. U.S. Fish Wild. Serv. FWS/OBS-80 no. 15.
- Hill, A.R. 1981. Stream phosphorus exports from watersheds with contrasting land uses in southern Ontario. *Water Resour. Bull.* 17: 627-634.
- Husar, R.B., T.J. Sullivan, and D.F. Charles. 1991. Methods in assessing long-term trends in atmospheric deposition and surface water chemistry. p. 65-82. *In* D.F. Charles (ed.) Acidic deposition and aquatic ecosystems: Regional case studies. Springer-Verlag New York, New York.
- Jokinen, E.H. 1983. Fresh water snails of Connecticut. State Geological and Natural History Survey of Connecticut. *Bull.* 109.
- Kahl, J.S., S.A. Norton, C.S. Cronan, I.J. Fernandez, L.C. Bacon, and T.A. Haines. 1991. Maine. p. 203-235. *In* D.F. Charles (ed.) Acidic deposition and aquatic ecosystems: Regional case studies. Springer-Verlag New York, New York.
- Kilham, P. 1982. Acid precipitation: Its role in the alkalization of a lake in Michigan. *Limnol. Oceanogr.* 27:856-867.
- Kramer, J., and A. Tessier. 1982. Acidification of aquatic systems: A critique of chemical approaches. *Environ. Sci. Technol.* 16: 606A-615A.
- Kramer, J.R., A.W. Andren, R.A. Smith, A.H. Johnson, R.B. Alexander, and G. Oehlert. 1986. Streams and lakes. p. 231-299. *In* Acid deposition: Long term trends. National Academy Press, Washington, DC.
- Krug, E.C., and C.R. Frink. 1983. Acid rain on acid soil: A new perspective. *Science* 221:520-525.
- Likens, G.E., F.H. Bormann, R.S. Pierce, J.S. Eaton, and N.M. Johnson. 1977. Biogeochemistry of a forested ecosystem. Springer-Verlag New York, New York.
- Marsicano, L.J., and P.A. Siver. 1993. A paleolimnological assessment of lake acidification in five Connecticut lakes. *J. Paleolimnol.* 9:209-221.
- Mattson, M.D., and P.J. Godfrey. 1994. Identification of road salt contamination using multiple regression and GIS. *Environ. Manage.* 18:767-773.
- Murray, T.E. 1994. Iron, sulfur, and phosphorus interactions in a softwater lake. Ph.D. thesis. Univ. of Connecticut, Storrs.
- Nilssen, S.I., H.G. Miller, and J.D. Miller. 1982. Forest growth as a possible cause of soil and water acidification: An examination of the concepts. *Oikos* 39:40-49.
- Nörusis, M.J. 1985. SPSSX: Advanced statistics guide. McGraw-Hill, New York.
- Norvell, W.A. 1977. Rapid changes in the composition of Linsley and Cedar ponds (North Branford, CT). *Arch. Hydrobiol.* 80:286-296.
- Norvell, W.A., and C.R. Frink. 1975. Water chemistry and fertility of twenty-three Connecticut lakes. *Connecticut Agric. Exp. Stn. Bull.* 759.
- Norvell, W.A., C.R. Frink, and D.E. Hill. 1979. Phosphorus in Connecticut lakes predicted by land use. *Proc. Natl. Acad. Sci.* 76:5426-5429.
- Omernik, J.M. 1976. The influence of land use on stream nutrient levels. USEPA Rep. 600/3-76-014. USEPA, Corvallis, OR.
- Osborne, L.L., and M.J. Wiley. 1988. Empirical relationships between land use/cover and stream water quality in an agricultural watershed. *J. Environ. Manage.* 26:9-27.
- Pionke, H.B., and J.B. Urban. 1984. Agricultural land use impacts on nutrients in ground water and base flow. p. 377-393. *In* Proc. National Water Well Assoc. Tech. Div., Eastern Regional Ground Water Conference, Newton, MA. 23-24 July 1984. Natl. Well Water Assoc., Worthington, OH.
- Pollman, C.D., and D.E. Canfield Jr. 1991. Florida. p. 237-271. *In* D.F. Charles (ed.) Acidic deposition and aquatic ecosystems: Regional case studies. Springer-Verlag New York, New York.
- Prowse, C.W. 1987. The impact of urbanization on major ion flux through catchments: A case study in southern England. *Water Air Soil Pollut.* 32:277-292.
- Renberg, I., T. Korsman, and H.J.B. Birks. 1993. Prehistoric increases in the pH of acid-sensitive Swedish lakes caused by land-use changes. *Nature (London)*362:824-826.
- Richter, D.D. 1984. Comment on acid precipitation in historical perspective and effects of acid precipitation. *Environ. Sci. Technol.* 18:632-634.
- Rogers, J.R., M. Gates, and J.L. Rosenfeld. 1959. Explanatory text for the preliminary geologic map of Connecticut, 1956. *Connecticut Geol. and Natural Hist. Surv. Bull.* 84.
- Rosenqvist, I.T. 1978. Acid precipitation and other possible sources for acidification of rivers and lakes. *Sci. Total Environ.* 10:271-272.
- Sisterson, D.L. 1991. Report 6. Deposition monitoring: Methods and results. p. 65-74. *In* P.M. Irving (ed.) Acid deposition: State of science and technology. Summary Rep. of the U.S. National Acid Precipitation Assessment Program, Washington, DC.
- Smol, J.P. 1992. Paleolimnology: An important tool for effective ecosystem management. *J. Aquat. Ecosyst. Health* 1:49-58.
- Smol, J.P., D.F. Charles, and D.R. Whitehead. 1984. Mallomonadacean microfossils provide evidence of recent lake acidification. *Nature (London)* 307:628-630.
- Stoddard, J.L., and P.S. Murdoch. 1991. Catskill Mountains. p. 237-271. *In* D.F. Charles (ed.) Acidic deposition and aquatic ecosystems: Regional case studies. Springer-Verlag New York, New York.
- Sullivan, T.J. 1991. Long-term temporal trends in surface water chemistry. p. 237-271. *In* D.F. Charles (ed.) Acidic deposition and aquatic ecosystems: Regional case studies. Springer-Verlag New York, New York.
- Sullivan, T.J., C.D. Driscoll, J.M. Eilers, and D.H. Landers. 1988. Evaluation of the role of sea salt inputs in the long-term acidification of coastal New England lakes. *Environ. Sci. Technol.* 22:185-190.
- Wagner, G.H., K.F. Steele, H.C. MacDonald, and T.L. Coughlin. 1976. Water quality as related to linears, rock chemistry and rain water chemistry in a rural carbonate terrain. *J. Environ. Qual.* 5: 444-451.
- Wanielista, M.P., Y.A. Yousef, and W.M. McLellon. 1977. Nonpoint source effects on water quality. *J. Water Pollut. Control Fed.* 49: 441-451.
- Weibel, S.R., R.J. Anderson, and R.L. Woodward. 1964. Urban land runoff as a factor in stream pollution. *J. Water Pollut. Control Fed.* 36:914-924.
- Wetzel, R.G., and G.E. Likens. 1991. Limnological analyses. Springer-Verlag New York, New York.