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Response of fish assemblages to declining acidic deposition in Adirondack Mountain lakes, 1984–2012

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HIGHLIGHTS

- Acidity of 43 long-term monitoring lakes has generally declined since the 1970s.
- Corresponding changes in community richness and fish catches were not evident.
- Fish metrics were weakly correlated with most acid-base chemistry parameters.
- Alternate monitoring plans are needed to quantify biological recovery in the region.
- Proactive stocking or liming could speed recovery of fisheries in Adirondack lakes.

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ABSTRACT

Adverse effects of acidic deposition on the chemistry and fish communities were evident in Adirondack Mountain lakes during the 1980s and 1990s. Fish assemblages and water chemistry in 43 Adirondack Long-Term Monitoring (ALTM) lakes were sampled by the Adirondack Lakes Survey Corporation and the New York State Department of Environmental Conservation during three periods (1984–87, 1994–2005, and 2008–12) to document regional impacts and potential biological recovery associated with the 1990 amendments to the 1963 Clean Air Act (CAA). We assessed standardized data from 43 lakes sampled during the three periods to quantify the response of fish-community richness, total fish abundance, and brook trout (*Salvelinus fontinalis)* abundance to declining acidity that resulted from changes in U.S. air-quality management between 1984 and 2012. During the 28-year period, mean acid neutralizing capacity (ANC) increased significantly from 3 to 30 μeq/L and mean inorganic monomeric Al concentrations decreased significantly from 2.22 to 0.66 μmol/L, yet mean species richness, all species or total catch per net night (CPNN), and brook trout CPNN did not change significantly in the 43 lakes. Regression analyses indicate that fishery metrics were not directly related to the degree of chemical recovery and that brook trout CPNN may actually have declined with increasing ANC. While the richness of fish communities increased with increasing ANC as anticipated in several Adirondack lakes, observed improvements in water quality associated with the CAA have generally failed to produce detectable shifts in fish assemblages within a large number of ALTM lakes. Additional time may simply be needed for biological recovery to progress, or else more proactive efforts may be necessary to restore natural fish assemblages in Adirondack lakes in which water chemistry is steadily recovering from acidification.

1. Introduction

Our understanding of the effects of acidic deposition on terrestrial and aquatic ecosystems has increased profoundly since the problem was first recognized approximately 50 years ago (Likens et al., 1972; Schofield, 1965). Watersheds across the southwestern Adirondack Mountains of New York commonly contain soils that are inherently low in base-cations (and the ability to neutralize inputs of strong acids) and also historically have received some of the highest inputs of acidic deposition in North

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America (Burns et al., 2011). As a consequence, soil and surface-water acidification from acidic deposition have caused chronic and episodic toxicity; and negatively affected biota in many lakes and streams across the region (Baker et al., 1996; Baldigo et al., 2007; Driscoll et al., 2001). With recognition of the adverse impacts on water quality and biology of surface waters in the Adirondacks and other acid-sensitive areas around the world (Beamish and Van Loon, 1977; Hesthagen, 1989; Rosseland et al., 1980), the U.S., Canada, and some European countries implemented regulations and policies such as the 1990 amendments to the U.S. Clean Air Act (CAA) of 1963 to lower rates of sulfur (S) and nitrogen (N) oxide emissions (Driscoll et al., 2010; Stoddard et al., 2003; Waller et al., 2012). While emissions of S peaked during the early to mid-1970s and N emissions peaked in the early 2000s, sulfate and nitrate concentrations in wet deposition declined by 74% and 65%, respectively, in the Adirondack region between 1980 and 2010 (Driscoll et al., this issue; Driscoll et al., 2001; Strock et al., 2014). Corresponding declines in mean concentrations of sulfate and nitrate (−47 μeq/L and −0.4 μeq/L, respectively) and increases in mean acid neutralizing capacity (ANC) (+12.6 μeq/L), evident in the Adirondack Long-Term Monitoring (ALTM) lakes of the Adirondack region from 1990 to 2010 (Stoddard et al., 2003; Strock et al., 2014), indicate the reductions in S and N emissions are improving the acid-base chemistry in surface waters (Waller et al., 2012). Additional declines in S and N emissions are also expected to be an indirect benefit of the 2015 Clean Power Plan which targets CO_2 emissions (Driscoll et al., *in review*).

Past research and monitoring of the chemical and biological responses to acidification in Adirondack lakes have been (and still are) critical to U.S. efforts to better understand the timing, severity, and extent of ecosystem impacts and develop emission targets for chemical recovery and the strategies to achieve proposed S and N emission targets (Greaver et al., 2012). In 1982, the ALTM program was initiated to characterize the acid-base chemistry of 17 Adirondack lakes on a monthly basis (Driscoll and Vandreason, 1993). This effort was followed by an extensive chemical and fishery survey of 1469 lakes conducted by the Adirondack Lakes Survey Corporation (ALSC), New York State Department of Environmental Conservation (NYSDEC), and others from 1984 to 1987 to ascertain the spatial extent and magnitude of acidification impacts throughout the Adirondack Region (Baker et al., 1990). Following this assessment, the original ALTM program was expanded in 1992 to monitor monthly chemistry (and conduct periodic fish resurveys) in 52 lakes (Fig. 1) across a broader range of lake sizes, drainage areas, and dissolved organic carbon (DOC) concentrations (Newton and Driscoll, 1990). Primary goals of the ALTM program were to document the responses of water chemistry and, where possible, fish assemblages to changing rates of acidic deposition in surface waters of the region and to improve our understanding of interrelated recovery processes (Roy et al., 2013).

Attempts to quantify changes or long-term trends in chemistry and biology of surface waters in the Adirondack region have been challenging because (a) the few research studies were not integrated; (b) continuous monitoring efforts largely focused on lake chemistry; and (c) biological assemblages were rarely quantified or monitored strategically. The effects of acidic deposition on acid-base chemistry and fish communities in Adirondack lakes (and to a lesser extent – streams), however, were documented during the 1980s and 90s (Baker et al., 1990, 1996; Colquhoun et al., 1984). Following the period of peak S and N emissions, surface-water concentrations of sulfate, nitrate, and inorganic monomeric Al_i have generally decreased, and the pH and ANC have generally increased significantly over time (Driscoll, 2011; Driscoll et al., *in review*; Lawrence et al., 2011; Stoddard et al., 2003; Waller et al., 2012). These results clearly indicate that implementation of the CAA and other emission regulations have improved water quality in lakes and streams of the Adirondacks. Unfortunately, few Adirondack lakes have long-term fishery data. The 1984–87 survey of 1469 lakes found that 346 (24%) were fishless and that 9 fish species (brook trout, *Salvelinus fontinalis*; white sucker, *Catostomus commersonii*; creek chub, *Semotilus atromaculatus*; common shiner, *Luxilus cornutus*; brown bullhead, *Ameiurus nebulosus*; pumpkinseed, *Lepomis gibbosus*; lake trout, *Salvelinus namaycush*; rainbow trout, *Oncorhynchus mykiss*; smallmouth bass, *Micropterus dolomieu*) and a sensitive-minnow group, were lost from numerous lakes probably due to acidification (Baker et al., 1990). Fish assemblages were resampled in all 52 ALTM lakes during 1994–2005 and again during 2008–12. While there has been a provisional analysis of changes in fish assemblages in 44 ALTM lakes between the first two survey periods (Roy et al., 2013), the occurrence frequencies for individual fish species in 43 ALTM lakes have not changed appreciably over time (Fig. S1) even though the acid sensitivity of each species differs greatly (Kretser et al., 1989). Because a comprehensive analysis of the potential changes in fish communities across the three survey periods has not been completed, the tangible effects of recent decreases in N and S emissions on biological recovery in Adirondack lakes – anticipated because of improvements in water quality across the region – remain unknown.

Whether recent regulations and declines in N and S emissions have stimulated recovery of biological systems in acidified lakes and streams of eastern North America is an important question for natural resource managers and for the promulgators of corresponding regulations. To address this question, the U.S. Geological Survey (USGS), NYSDEC, Syracuse University, and the New York State Energy Research and Development Authority initiated the present study. The main goal of this investigation was to evaluate if recent changes in acid-base chemistry have significantly affected fish assemblages in acidified lakes of the Adirondack region between 1984 and 2012 using existing data collected under the ALTM program. Our findings will have important implications for management of fisheries and water quality in lakes of the region. First, this work can be used to help define (or refine) chemistry-fish response models and detect chemical thresholds that limit the occurrence of selected indicator species or species groups. Second, the value of the original (1984–87) fisheries dataset and both resurveys will be assessed; i.e., results will help project and validate anticipated recovery of lake ecosystems and develop the strategies needed to more effectively monitor long-term changes in fish assemblages. Third, study results will apprise the need for additional remedial efforts to accelerate the chemical and biological recovery of previously acidified lakes. Lastly, our results will directly inform decisions regarding the new (impending) secondary standards for nitrogen dioxide (NO_2) and sulfur dioxide (SO_2) and how fish resources may be adaptively managed in lakes across the Adirondack region.

2. Materials and methods

Large sets of ALTM lake chemistry and fishery data had to be compiled, standardized, and integrated for intended analyses. First, fish and chemistry data from the three surveys were acquired, standardized, and filtered (to exclude sites without comparable data from all 3 periods and records for stocked fish species). Second, acid-base chemistry (e.g., ANC, pH, inorganic monomeric Al) and fish community metrics (species richness and all species or total catch per net night (CPNN)) and brook trout CPNN were compiled for each lake and survey date. Third, the relations between the three fish-community metrics and ANC, Al_i, and pH were explored using regression analysis to predict potential temporal shifts in

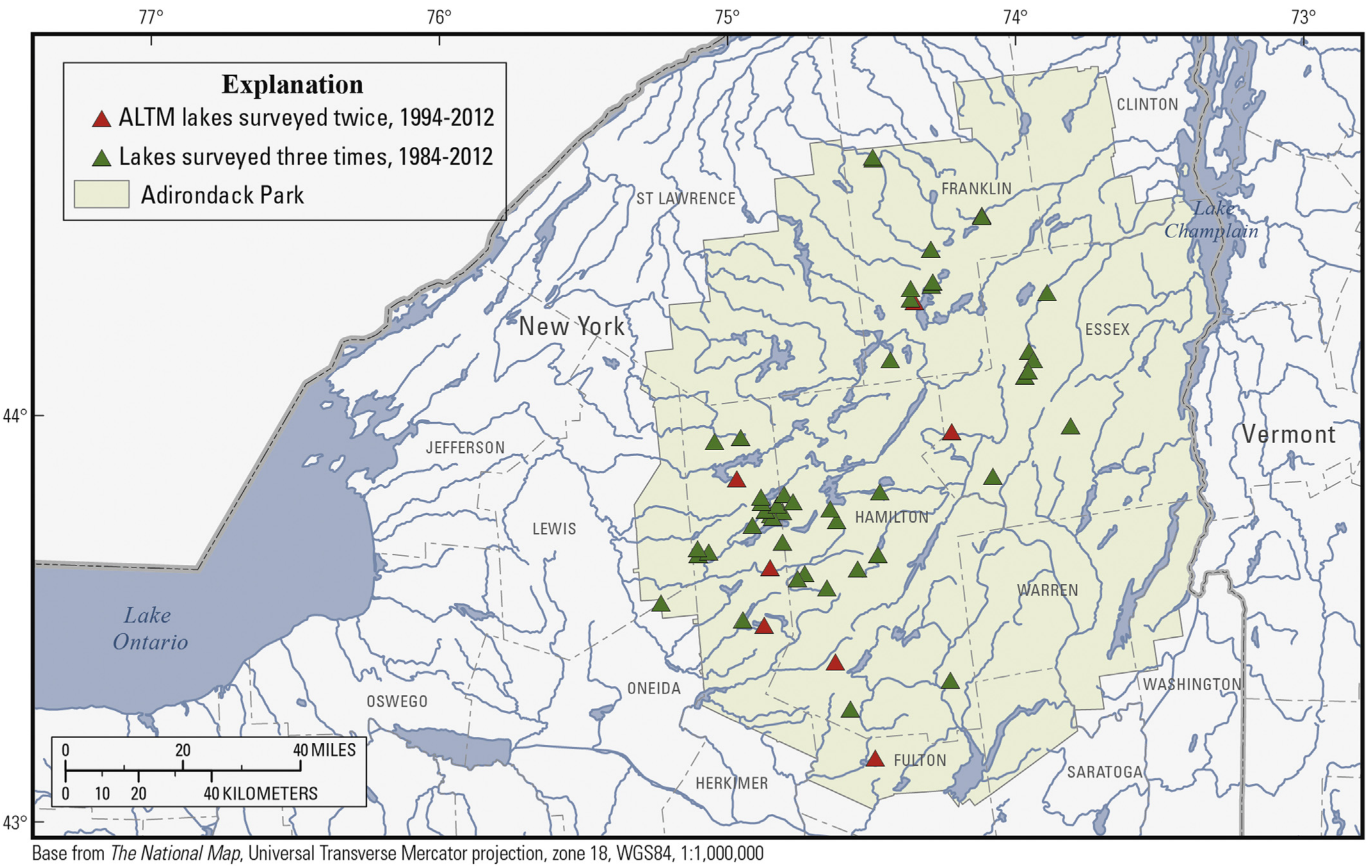

Fig. 1. Location of 43 Adirondack Long-Term Monitoring (ALTM) Lakes where fish surveys were completed during 1984–87, 1994–2005, and 2008–12 (green triangles). The lakes where fish assemblages were only surveyed during the second and third periods are denoted by red triangles. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

metrics that could be associated with changes in chemistry. Fourth, the distributions of acid-base chemistry parameters from all lakes, with analogous data from all 3 periods, were assessed to determine if temporal changes were significant. Fifth, the distributions of fishery metrics from the study lakes were assessed to determine if temporal changes were evident and significant. The study area, fish-survey methods, and analytical procedures are described in more detail below.

2.1. Study area and lakes

The study area is entirely contained within the boundaries of the Adirondack Park – a large, mostly forested upland area of 4585 km^2 in northern New York State. Characteristics of the region are reported in numerous publications including Roy et al. (2013), Lawrence et al. (2008), Baker et al. (1990), Driscoll and Newton (1985), and Driscoll et al. (1991). Briefly, the irregular terrain in this mountainous region was repeatedly glaciated leaving many bedrock-scoured ridgetops. The surface areas of ALTM lakes range from 1.2 to 187 ha and lake elevations range from 381 to 873 m. The region is generally classified as northern hardwoods; red spruce and balsam fir are often dominant at higher elevations. Extensive wetlands are common in many watersheds. Hillside surficial deposits are relatively thin, and drift on valley floors can be up to hundreds of meters deep. Bedrock geology is a complex mixture of granitic and gneissic rocks, interspersed with other less common metasedimentary formations; surficial deposits mirror the bedrock geology and include highly weatherable calcareous minerals in certain areas.

Mean annual precipitation across the region ranges from about 800 to over 1600 mm (Ito et al., 2002). The pH of precipitation averaged about 4.1 during the 1980s (Baker et al., 1990). During 2013, the pH of precipitation averaged about 5.0, yet sulfate and nitrate (acidic deposition) concentrations remained among the highest in the Northeast (NADP, 2014). Long periods of below-freezing temperatures in winter typically result in deep accumulations of snow, which melt rapidly in the spring causing sustained high stream flows (Lawrence et al., 2008). The ionic strength of most surface waters across western and southwestern parts of the Park remain especially dilute, with low levels of ANC.

The ALTM lakes are distributed across the Adirondack Park (Fig. 1) and vary widely in elevation, watershed area, lake area, and sensitivity to acidic deposition (Table 1). Only 43 of 52 ALTM lakes had analogous fish-survey data from all three survey periods. Little Simon Pond, Little Echo Pond, Woods Lake, Big Moose Lake, South Lake, Arbutus Lake, Otter Lake, and G Lake were not included in the 1984–87 survey and, therefore, were not included in our analyses. An additional lake, Black Pond (30256), was also excluded because it was reclaimed one year before its second survey. Because lake sensitivities (and responses) to acidic deposition varied widely, a classification system for acid-base status, based primarily on surficial geology and hydrologic flow paths for each lake, was devised in the late 1980s (Table 1) (Baker et al., 1990; Driscoll et al., 2003). Wetlands within lake watersheds may also raise levels of naturally occurring organic acids which affect acidity and toxicity of Al_i; thus, ALTM lakes were also classified as having high or low DOC concentrations (greater or less than 500 μmol C/L, respectively) as noted in Table 1. The NYSDEC actively manages brook trout, and a

Table 1
The ID, name, location, and characteristics of 43 Adirondack Long Term Monitoring lakes sampled during three periods, 1984–87, 1994–05, and 2008–12. [DOC, dissolved organic carbon].

Lake ID	Lake/Pond name	Latitude	Longitude	Elevation (m)	Max depth (m)	Mean depth (m)	Volume (10^4 m^3)	Surface area (ha)	Water shed area (ha)	Water shed ratio	Hydraulic retention (years)	Lake classification	DOC class
020058	LITTLE HOPE POND	44° 30′ 57″	−074° 07′ 31″	517	6.2	3.5	10.0	2.8	53.6	0.05	0.29	Medium Till Drainage	High
020059	BIG HOPE POND	44° 30′ 43″	−074° 07′ 30″	517	11.5	5.8	51.6	8.9	119.2	0.07	0.68	Medium Till Drainage	High
020138	EAST COPPERAS POND	44° 18′ 43″	−074° 22′ 20″	480	6.4	4.1	14.8	3.6	13.0	0.28	1.78	Thin Till Drainage	High
020143	MIDDLE POND	44° 20′ 13″	−074° 22′ 19″	483	3.3	1.5	36.9	24.3	187.1	0.13	0.31	Carbonate Influenced	High
020188	SUNDAY POND	44° 20′ 41″	−074° 18′ 02″	495	11.0	5.4	21.9	4.0	21.5	0.18	NA	Mounded Seepage	Low
020197	SOCHIA POND	44° 21′ 08″	−074° 17′ 41″	495	5.5	3.1	5.0	1.6	9.6	0.17	NA	Mounded Seepage	Low
020233	OWEN POND	44° 19′ 23″	073° 54′ 12″	514	9.4	3.7	28.4	7.6	1159.0	0.01	0.03	Thick Till Drainage	Low
020264	HEART LAKE	44° 10′ 47″	073° 58′ 03″	661	16.8	5.1	54.5	10.7	69.3	0.15	1.03	Medium Till Drainage	Low
020265	MARCY DAM POND	44° 09′ 32″	073° 57′ 11″	720	2.4	0.7	0.8	1.2	1177.2	0.00	0.00	Thin Till Drainage	Low
030171	GRASS POND	44° 39′ 26″	−074° 29′ 45″	381	7.0	4.2	6.8	1.6	29.5	0.05	NA	Mounded Seepage	High
030172	LITTLE CLEAR POND	44° 39′ 38″	−074° 29′ 53″	381	14.0	5.5	10.2	1.9	18.0	0.11	NA	Mounded Seepage	Low
040186	LOON HOLLOW POND	43° 57′ 41″	−075° 02′ 43″	605	11.6	3.4	19.1	5.7	55.2	0.10	0.46	Thin Till Drainage	Low
040210	WILLYS LAKE	43° 58′ 20″	−074° 57′ 20″	632	13.7	4.9	118.8	24.3	158.2	0.15	1.00	Thin Till Drainage	Low
040704	MIDDLE SETTLEMENT LAKE	43° 41′ 02″	−075° 06′ 00″	526	11.0	3.4	54.5	15.8	114.3	0.14	0.63	Thin Till Drainage	Low
040706	GRASS POND	43° 41′ 25″	−075° 03′ 54″	549	5.2	1.5	7.8	5.3	272.0	0.02	0.04	Medium Till Drainage	Low
040707	MIDDLE BRANCH LAKE	43° 41′ 52″	−075° 06′ 08″	496	5.2	2.1	36.3	17.0	129.6	0.13	0.37	Thin Till Drainage	Low
040739	LAKE RONDAXE	43° 45′ 23″	−074° 54′ 59″	524	10.1	3.0	273.3	90.5	14155.6	0.01	0.03	Thin Till Drainage	Low
040746	MOSS LAKE	43° 46′ 52″	−074° 51′ 11″	536	15.2	5.7	259.8	45.7	1234.6	0.04	0.28	Medium Till Drainage	Low
040748	BUBB LAKE SSTA1	43° 46′ 29″	−074° 50′ 49″	554	4.3	2.1	38.5	18.2	199.1	0.09	0.25	Thin Till Drainage	Low
040750	DART LAKE	43° 47′ 36″	−074° 52′ 16″	537	17.7	7.3	380.7	51.8	10804.5	0.01	0.05	Thin Till Drainage	Low
040750A	WINDFALL POND SSTA1	43° 48′ 18″	−074° 49′ 53″	591	6.1	3.2	7.8	2.4	41.1	0.06	0.25	Carbonate Influenced	Low
040753	WEST POND SSTA1	43° 48′ 41″	−074° 53′ 00″	581	5.2	1.5	15.2	10.4	99.6	0.10	0.20	Thin Till Drainage	Low
040754	SQUASH POND SSTA1	43° 49′ 32″	−074° 53′ 11″	653	5.8	1.4	4.5	3.3	125.1	0.03	0.05	Thin Till Drainage	High
040777	CONSTABLE PD SSTA1	43° 49′ 50″	−074° 48′ 27″	580	4.0	2.1	43.5	20.6	937.4	0.02	0.06	Thin Till Drainage	Low
040826	LIMEKILN LAKE	43° 42′ 48″	−074° 48′ 47″	575	21.9	6.1	1147.6	186.9	1409.7	0.13	1.07	Medium Till Drainage	Low
040850	SQUAW LAKE	43° 38′ 10″	−074° 44′ 20″	646	6.7	3.4	124.9	36.4	182.7	0.20	0.77	Thin Till Drainage	Low
040852	INDIAN LAKE	43° 37′ 24″	−074° 45′ 44″	654	10.7	3.0	98.1	33.2	1121.4	0.03	0.10	Thin Till Drainage	Low
040874	BROOK TROUT LAKE	43° 36′ 00″	−074° 39′ 45″	724	23.2	8.4	242.0	28.7	165.7	0.17	1.64	Thin Till Drainage	Low
040887	LOST POND	43° 38′ 48″	−074° 33′ 30″	717	1.2	0.7	3.2	4.4	173.8	0.03	0.03	Thin Till Drainage	High
040905	BARNES LAKE	43° 33′ 52″	−075° 13′ 36″	395	10.1	4.5	13.1	2.9	6.5	0.45	NA	Mounded Seepage	High
041007	NORTH LAKE	43° 31′ 22″	−074° 56′ 53″	555	17.7	5.7	1010.7	176.8	7700.8	0.02	0.15	Thin Till Drainage	Low
050215	WILLIS LAKE	43° 22′ 17″	−074° 14′ 47″	400	2.7	1.6	22.9	14.6	136.4	0.11	0.22	Medium Till Drainage	Low
050259	JOCKEYBUSH LAKE	43° 18′ 08″	−074° 35′ 09″	599	11.3	4.5	78.6	17.3	160.0	0.11	0.55	Thin Till Drainage	Low
050458	CLEAR POND	43° 59′ 38″	073° 49′ 40″	584	24.4	9.2	651.1	70.4	565.0	0.12	1.80	Thick Till Drainage	Low
050577	NATE POND	43° 52′ 23″	−074° 05′ 45″	613	6.4	2.3	19.4	8.3	89.2	0.09	0.29	Medium Till Drainage	High
050649	LONG POND	43° 50′ 15″	−074° 28′ 50″	574	4.0	2.0	3.3	1.7	29.6	0.06	0.15	Thin Till Drainage	High
050669	CARRY POND	43° 40′ 54″	−074° 29′ 21″	652	4.6	2.2	6.2	2.8	20.8	0.13	NA	Mounded Seepage	Low
050706	LAKE COLDEN	44°07′ 09″	073° 58′ 59″	843	7.3	2.3	35.5	15.4	656.3	0.02	0.08	Thin Till Drainage	Low
050707	AVALANCHE LAKE	44° 07′ 51″	073° 58′ 13″	873	7.0	3.3	14.6	4.4	115.2	0.04	0.20	Thin Till Drainage	Low
060182	LITTLE SIMON POND	44° 09′ 42″	−074° 26′ 36″	546	32.0	11.0	631.3	58.1	774.0	0.08	1.28	Medium Till Drainage	Low
060313	SAGAMORE LAKE	43° 45′ 57″	−074° 37′ 43″	580	22.9	10.5	713.1	68.0	4723.0	0.01	0.20	Medium Till Drainage	High
060315A	RAQUETTE LAKE RES	43° 47′ 42″	−074° 39′ 05″	564	3.0	1.6	2.4	1.5	305.5	0.01	0.01	Medium Till Drainage	High
060329	QUEER LAKE	43° 48′ 49″	−074° 46′ 38″	597	21.3	10.9	596.0	54.5	375.4	0.15	2.10	Thin Till Drainage	Low

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few other fish species, in about half of these lakes with annual or intermittent stockings (Roy et al., 2011). Additional details about the study area and individual ALTM lakes can be found in Driscoll et al. (2003) and Roy et al. (2011).

2.2. Fish surveys

Fish assemblages were inventoried in 43 ALTM lakes during 1984–87, and in 52 ALTM lakes during 1994–2005 and 2008–12, following standard ALSC methods (Baker et al., 1990). Briefly, fish assemblages in each lake were inventoried once in either the spring (April–June) or fall (September–November) between 1984 and 1987. Several types of gear were employed to best document the diversity of fish species in each lake. Experimental Swedish gill nets (45.7 m long), with 5 panels and 38–90 mm (stretch) mesh were the primary sampling gear. The number of nets set per lake varied with surface area; 0 nets for lakes <4.0 ha, 2 nets for lakes 4.1–10.1 ha, 3 nets for lakes 10.2–20.2 ha, 4 nets for lakes 20.3–40.5 ha, and 4 plus 1 additional net per 40 ha for lakes larger than 40.5 ha. These nets were usually set perpendicular to shore, on the lake bottom, and across depth contours to or towards the maximum lake depth – as long as dissolved oxygen (DO) was adequate for fish survival. Nets were also set parallel to shore, at constant depth, above the depth where DO was not adequate for fish survival. Where minimal fish mortality was requisite, modified Alaska or Oneida trap nets were used in place of, or along with, the experimental gill nets. The number of trap nets per lake; however, was increased by 1 over the number suggested for gill nets. Additionally, at least one minnow trap and one, 9.2-m long monofilament or multifilament gill net with 1.9-mm (stretch) mesh were set in the littoral zone of each lake to sample juvenile fish and small minnow species. All gear was typically set overnight (15–24 h deployments), but sometimes retrieved sooner to reduce mortality, predation, or overcrowding. The number of each species captured was recorded and total lengths and weights from a subsample of as many as 20 individuals (from representative length classes) of each species were noted. This protocol was followed during the first and third (2008–12) surveys, but the sampling effort was augmented (additional net sets and additional net types were used) in many lakes during the second (1994–2005) survey (Roy et al., 2011). Fish netting summaries from the first (1984–87) survey of most ALTM lakes are available in Roy et al. (2011) and can be searched at: "http://www.adirondacklakessurvey.org/historic.php". Fish community metrics and CPNN data for brook trout in the 43 lakes sampled during all three survey periods are summarized in Table S1.

2.3. Lake chemistry

Water chemistry data were obtained from samples collected at 1.5 m below the surface at the deepest part or approximate middle of each lake during the original ALSC survey of 1469 lakes and the ongoing ALTM program. During the first period (1984–87), water chemistry of the 43 ALTM lakes were determined from two samples; one collected during fish surveys in either the spring or fall, and a second collected during July or August of the same year that fish were sampled (Baker et al., 1990). Because chemistry of each lake was assessed twice (once during summer and once either in fall or spring), and chemistry can change considerably across seasons, the summer data were used to represent the most steady (baseline) condition and to evaluate temporal changes in water quality. Water samples also have been collected monthly from all 52 ALTM lakes from 1992 to 2012. Chemistry data corresponding to the second and third fish surveys were obtained from the sample collected closest (generally within 2 weeks) to the date that the first summer (1984–87) baseline sample was collected. All water samples were analyzed for 30 physio-chemical parameters (Baker et al., 1990; Roy et al., 2013) and included important acid-base constituents such as pH, ANC, and Al_i. Concentrations of Al_i from samples collected during the 1984–87 survey were not measured directly, but estimated from measured total Al, pH, and DOC using a chemical equilibrium model (Baker et al., 1990; Fakhraei and Driscoll, 2015). Another acid-base chemistry indicator, base-cation surplus (BCS), was also estimated for most samples following methods as described in Lawrence et al. (2007). Base-cation surplus accounts for the effects of natural organic acidity on the acid-neutralizing capacity of surface waters. Below a BCS value of zero, toxic Al_i is mobilized (due to increased acidity), whereas above a BCS of zero, base cation concentrations exceed the total concentrations of strong acid anions (including strongly acidic organic anions), and Al_i is not mobilized. Summer chemistry data from the 1984–87 survey of 43 ALTM lakes are provided in Table S2 and can be searched at: "http://www.adirondacklakessurvey.org/historic.php". Monthly chemistry data for the 43 ALTM lakes are available at "http://www.adirondacklakessurvey.org/".

2.4. Data analyses

Chemistry and fishery data obtained from all 43 lakes during each survey period were compiled separately and then merged for analysis. Data for important acid-base chemistry variables from the 43 ALTM lakes sampled during all three periods were compiled into one spreadsheet and sorted by survey period, year, and lake ID (Table S2). Fishery data were standardized to only include information collected using the same level of effort and the same gear types across all three survey periods. The increased number of net sets (of the same gear type) used in most lakes during the second survey and the variable number of nets (of the same or different gear type), deployed across all ALTM lakes required that species counts be divided by the total number of nets (of the same gear type) used in each lake to standardize catch per unit effort (CPUE) as catch per net night (CPNN). Because the number of individuals and species captured is directly related to sampling effort (Gotelli and Colwell, 2001), additional sets of the same gear type at many lakes likely biased (increased) species richness during the second survey. More important, all data collected using non-standard gear types (e.g., Alaska trap, beach seines) during the second (and sometimes third) survey in many lakes had to be removed (filtered) from all analyses to minimize bias in species richness. Once standardized and filtered, the CPNN data for individual species were calculated (number of each species divided by the total number of standard gear types deployed in each lake), along with total CPNN (all fish species combined) and species richness, from each of the three surveys. Lastly, unless known to be self-sustaining, observations of fish species that were stocked by the NYSDEC within 5 years prior to any fish-survey period, were excluded from the dataset to remove the effects of management actions on our analysis of naturally recovering fish assemblages.

Key chemistry data and fishery metrics were merged into one dataset by survey period, year, and lake to conduct two discrete analyses. First, the chemistry data were analyzed to determine if: (a) ANC, pH, BCS, DOC, Calcium (Ca), or Al_i from all 43 lakes differed significantly among the three survey periods using repeated measures ANOVA on ranks and Tukey's test of multiple comparisons; (b) the magnitude of change in chemical constituents differed significantly among periods; and (c) the distribution of measures for each constituent from the 43 lakes differed significantly among the three survey periods using two-sample Kolmogorov-Smirnov (KS) tests. Second, the relations between key fishery metrics and ANC, pH, BCS, or Al_i were analyzed using linear regression to: (a) identify the strength of relations; (b) explore/detect any potential effect thresholds; and (c) predict the potential change in fishery metrics expected with chemistry that changed significantly over the three periods. All analyses were used to test the null hypothesis that selected chemical parameters and brook trout CPNN, community richness, or total CPNN did not change significantly at all repeatedly sampled sites among the three survey periods.

3. Results

3.1. Lake chemistry

Though not the primary focus of this paper, significant changes in the acid-base chemistry of the ALTM lakes had to be established before the response of fish assemblages could be inferred (hypothesized) and the expected changes assessed. Our analysis of temporal changes in acid-base chemistry of the 43 study lakes confirmed that acidity and Al_i concentrations (and water toxicity) declined significantly between at least the first and third surveys. The ANC levels increased in 30 (70%) of the 43 lakes between the first and second survey, and in 38 (88%) of the lakes between the first and third survey (Table S2). The ANC of all samples collected from the 43 ALTM lakes during summer ranged from −45.8 to 149.2 μeq/L (Table S2) across the three periods and the mean ANC increased significantly from the first (17.7 μeq/L) to second (26.2 μeq/L) surveys, and from the first to third (37.7 μeq/L) surveys (Table 2, Fig. 2A). Of greater importance to biota, Al_i concentrations decreased in 32 (74%) and 41 (95%) of the 43 lakes between the first and the second and third surveys, respectively (Table S2). Mean Al_i concentrations decreased significantly between the first (2.22 μmol/L) and second (1.55 μmol/L), and between the first and third (0.66 μmol/L), surveys (Table 2, Fig. 2D). Likewise, estimates of mean BCS differed significantly from the first (5.6 μeq/L) to second (12.3 μeq/L) survey and from the first to third (29.6 μeq/L) survey (Table 2). Increases in mean pH and DOC concentrations were only significant between the first and third surveys (Table 2, Fig. 2B, C), whereas, mean calcium (Ca) concentrations decreased significantly between the first and second, and between the first and third, surveys (Table 2, Fig. 2E).

The distributions of ANC and Al_i data from all 43 ALTM lakes differed significantly across the three survey periods, which substantiate observed shifts in their measures of central tendency and suggest that the acid-base status of Adirondack lakes is improving on a broad scale. The distributions of ANC data (Fig. 3A) from the three surveys suggest that the median increased by approximately 25 μeq/L between the first and third surveys. Two-sample KS tests indicate that the cumulative frequency distributions for ANC differed significantly ($P = 0.0307$) between the first and third surveys, but not between the first and second surveys (Table 2, Fig. 3B). The KS tests indicate that the cumulative frequency distributions for Al_i differed significantly between both the first and third surveys ($P < 0.0001$) and between the first and second surveys ($P = 0.0003$) (Table 2, Fig. 4B). The cumulative frequency distributions for the other chemistry variables did not differ significantly among the three survey periods, yet differences in the distribution of BCS, Ca, and pH data would be considered marginally significant at $P = 0.10$ between the first and third surveys (Table 2).

3.2. Fish assemblages

At the largest scale (all 43 lakes), the number of species in fish communities did not respond to significant improvements in acid-base chemistry between the first and second or between the first and third surveys. Mean richness averaged 2.9, 3.4, and 3.1 species in all 43 lakes during the first, second, and third surveys, respectively, and did not differ significantly among periods (Table 2, Fig. 5A). The two-sample KS tests confirmed that cumulative frequency distributions (Fig. 6B) for species richness data from all 43 ALTM lakes (Fig. 6A) did not differ significantly among any of the surveys (Table 2). At a smaller (individual lake) scale, however, species richness was higher in 18 (42%) of the individual lakes during the second survey, and in 14 (33%) of the lakes during the third survey as compared to the first survey (Table S1). Although the number of fish species collected from individual lakes was highly variable through time, increases in the number of fish species in at least one-third of the ALTM lakes between 1984–87 and 2008–12 may be important to regional aquatic ecosystems.

Table 2
Results (*P*-values) from two-sample Kolmogorov-Smirnov tests, Shapiro-Wilks normality tests, Brown-Forsyth equal-variance tests, and Tukey's multiple comparison tests for repeated measures ANOVA on ranks describing the significance of differences in mean acid neutralizing capacity (ANC), pH, calcium, inorganic Al, dissolved organic carbon (DOC), base-cation surplus (BCS), and fishery metrics or their distributions in 43 ALTM lakes sampled three times: 1984–87 (Survey 1, S1), 1994–2005 (Survey 2, S2), and 2008–12 (Survey 3, S3). Mean values with the same superscript letter for a given parameter do not differ significantly based on the repeated measures ANOVA on ranks and Tukey's multiple comparison tests; *P*-values less than 0.05 are denoted in bold.

Parameter	Survey 1 mean	Survey 2 mean	Survey 3 mean	Normality test *P*-value	Equal variance test *P*-value	Repeated measures ANOVA *P*-value	Tukey's multiple comparison test			Two-sample Kolmogorov-Smirnov test		
							P-value S1-S2	*P*-value S1-S3	*P*-value S2-S3	*P*-value S1-S2	*P*-value S1-S3	*P*-value S2-S3
Acid-base chemistry												
ANC (μeq/L)	17.7^{a}	26.2^{b}	37.7^{c}	**<0.05**	0.175	**<0.001**	**0.023**	**<0.001**	**0.001**	0.269	**0.031**	0.407
pH	5.58^{a}	5.67^{a}	5.92^{b}	**<0.05**	0.274	**<0.001**	0.356	**<0.001**	**<0.001**	0.917	0.101	0.169
Calcium (μmol/L)	44.5^{a}	39.7^{b}	35.8^{c}	**<0.05**	0.273	**<0.001**	**0.001**	**<0.001**	**0.010**	0.269	0.101	0.580
Inorganic Al (μmol/L)	2.22^{a}	1.55^{b}	0.66^{c}	**<0.05**	0.434	**<0.001**	**0.005**	**<0.001**	**<0.001**	**<0.001**	**<0.001**	0.057
DOC (μmol C/L)	415.7^{a}	448.2^{ab}	494.8^{b}	**<0.05**	0.480	**0.002**	0.244	**0.002**	0.115	0.917	0.169	0.407
BCS (μeq/L)	5.6^{a}	12.3^{b}	29.6^{b}	0.051	0.559	**<0.001**	**<0.001**	**<0.001**	0.122	0.765	0.101	0.057
Fishery metrics												
Richness (No. of species)	2.9^{a}	3.4^{a}	3.1^{a}	**<0.05**	0.934	0.141	0.141	0.141	0.141	0.141	0.141	0.141
All species catch per net night	16.0^{a}	22.0^{a}	14.5^{a}	**<0.05**	0.866	0.238	0.238	0.238	0.238	0.238	0.238	0.238
Brook trout catch per net night	0.69^{a}	0.60^{a}	0.68^{a}	**<0.05**	0.946	0.962	0.962	0.962	0.962	0.962	0.962	0.962

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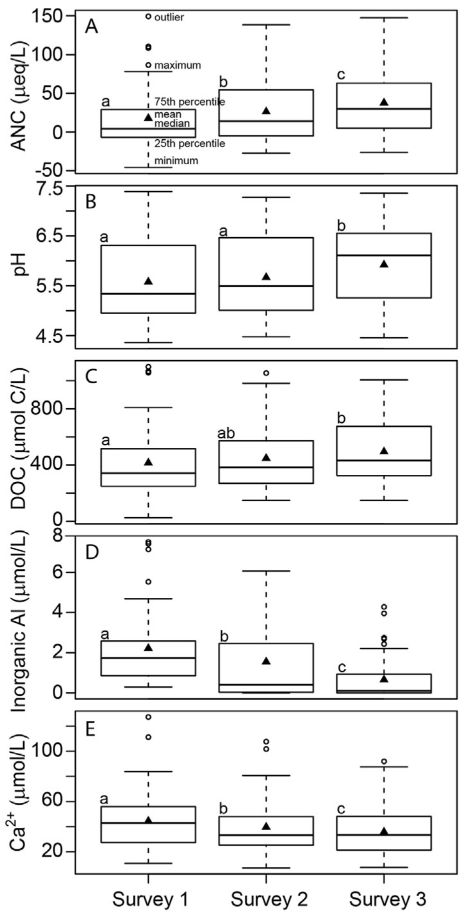

Fig. 2. Mean, median, 25th to 75th percentiles, minimum, maximum, and outlier values for (A) acid neutralizing capacity (ANC), (B), pH, (C) dissolved organic carbon (DOC), (D) inorganic aluminum (Al_i), and (E) calcium (Ca) from water samples collected in 43 ALTM lakes once during July or August of the same year that fish were sampled during three survey periods (1984–87, 1994–2005, and 2008–12). Different letters denote significant differences among the three survey periods determined using repeated measures ANOVA on ranks and Tukey's multiple comparisons tests.

Total CPNN from the 43 ALTM lakes, like richness, did not respond to significant improvements in acid-base chemistry between the first and second or third surveys. Mean CPNN averaged 16.1, 22.0, and 14.5 fish per net night in all 43 study lakes during the first, second, and third surveys, respectively, and did not differ significantly among periods (Table 2, Fig. 5B). The two-sample KS tests showed that cumulative frequency distributions (Fig. 7B) for all species CPNN data from all study lakes (Fig. 7A) did not differ significantly among the three surveys (Table 2). Total CPNN was also higher in 16 (37%) of the lakes during the second survey, and in 13 (30%) of the lakes during the third survey as compared to the first survey (Table S1).

The brook trout CPNN also did not respond to significant changes in acid-base chemistry between the first and second or first and third surveys in the 43 ALTM lakes. Mean brook trout CPNN was 0.69, 0.60, and 0.68 fish in the 43 study lakes during the first, second, and third surveys, respectively, and did not differ significantly among surveys (Table 2, Fig. 5C). The two-sample KS tests also showed that cumulative frequency distributions (Fig. 8B) for brook trout CPNN (Fig. 8A) did not differ significantly between the three surveys (Table 2). Brook trout CPNN was also higher in 5 (12%) of the 43 lakes during the second survey, and in 8 (19%) of the 43 lakes during the third survey, as compared to the first survey (Table S1).

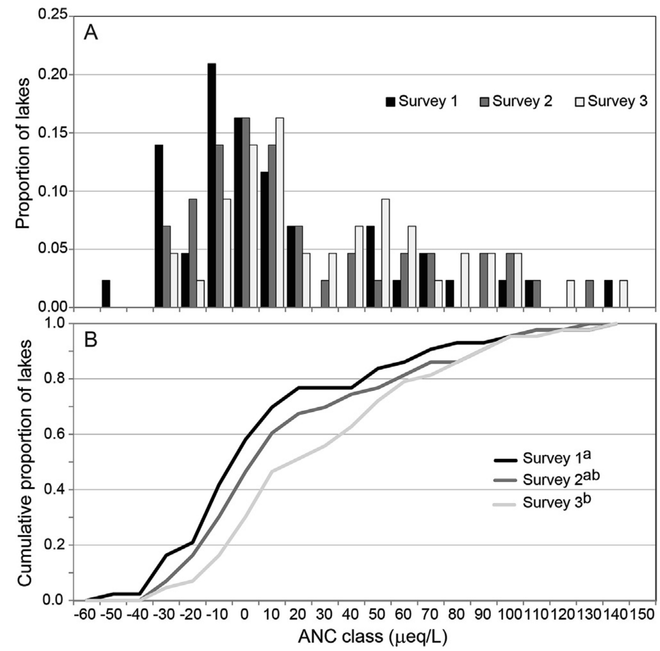

Fig. 3. The (A) proportion (frequency distribution) and (B) the cumulative proportion of lakes in 10 μeq/L acid neutralizing capacity (ANC) classes from water samples collected in 43 ALTM lakes once during July or August of the same year that fish were sampled during three survey periods (1984–87, 1994–2005, and 2008–12). Different letters denote significant differences in the lake ANC distributions among the three survey periods using two-sample Kolmogorov-Smirnov tests.

3.3. Fish and chemistry relations

An assessment (or validation) of the relations between acid-base chemistry and key fishery metrics in our study lakes is

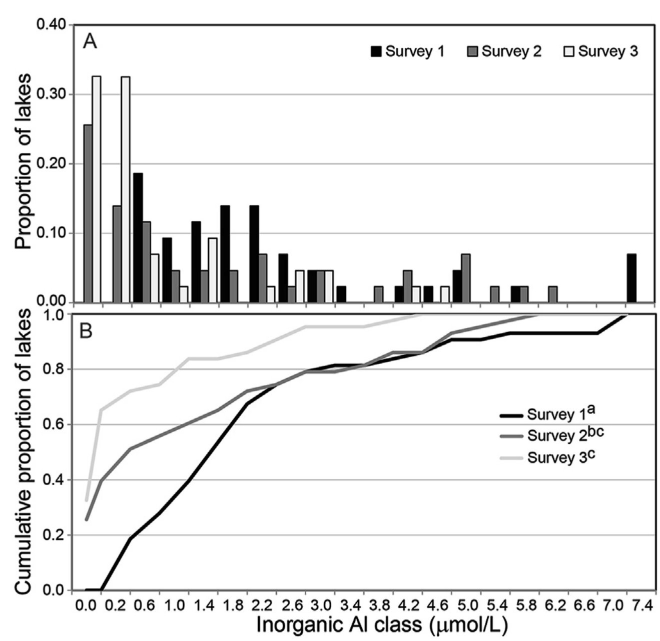

Fig. 4. The (A) proportion (frequency distribution) and (B) the cumulative proportion of lakes in 0.4 μmol/L inorganic aluminum (Al_i) classes from water samples collected in 43 ALTM lakes once during July or August of the same year that fish were sampled during three survey periods (1984–87, 1994–2005, and 2008–12). Different letters denote significant differences in the lake inorganic aluminum (Al_i) distributions among the three survey periods using two-sample Kolmogorov-Smirnov tests.

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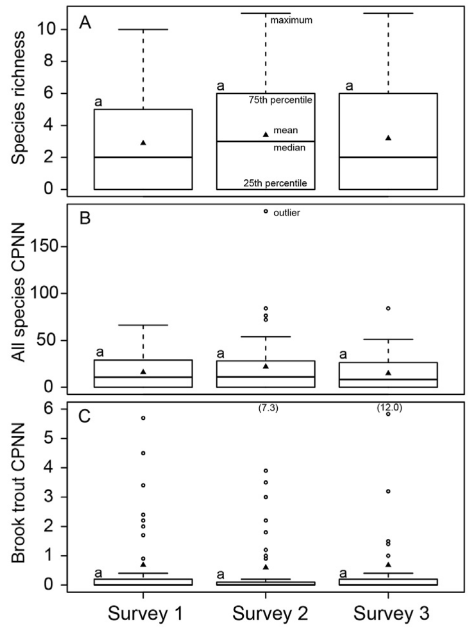

Fig. 5. Mean, median, 25th to 75th percentiles, minimum, maximum, and outlier values for (A) total species richness, (B) all fish species or total catch per net night (CPNN), and (C) brook trout CPNN in 43 ALTM lakes surveyed during three periods (1984–87, 1994–2005, and 2008–12). Different letters denote significant differences among the three survey periods using repeated measures ANOVA on ranks and Tukey's multiple comparisons tests.

useful because resident fish populations and communities would only be expected to recover with improving water quality if they were either directly or indirectly related. The results of simple (linear) regression analyses across periods identified significant and moderately strong relations (R^2 values range from 0.11 to 0.41) between species richness and ANC (Fig. 9A), Al_i (Fig. 9B), and pH levels (Fig. 9C). Notably, relatively few or no fish species were collected from lakes with summertime ANC values less than 0 μeq/L, pH levels less than 5.0, and Al_i concentrations greater than 4 μmol/L. Although not as strong (R^2 values range from 0.07 to 0.21), significant relations were also evident between all species CPNN and ANC (Fig. 9D), Al_i (Fig. 9E), and pH levels (Fig. 9F). Total fish CPNN, like richness, was generally reduced or zero in lakes with summertime ANC values less than 0 μeq/L, pH levels less than 5.0, and Al_i concentrations greater than 4 μmol/L. There was no significant linear relation between brook trout CPNN and any acid-base chemistry variable (Fig. 9), yet brook trout CPNN was very low or zero in lakes with ANC less than 0 μeq/L or greater than 70 μeq/L (Fig. 9G), pH levels less than 5.0 or greater than 6.5 (Fig. 9I), and Al_i concentrations greater than 4 μmol/L (Fig. 9H). These spatial relations between chemical characteristics and fish metrics coupled with significant temporal improvements in acid-base chemistry generally support the hypothesis that biological recovery should be evident in many ALTM lakes.

4. Discussion

Though our analysis of changes in water chemistry was limited, the significant increases in pH and ANC, and decreases in Al_i concentrations, were critical for interpreting fishery results because they demonstrate that water quality of study lakes improved along with concomitant decreases in acid deposition over the past three decades. The extensive changes in acid-base chemistry of the 43 ALTM lakes noted between the first and third survey periods generally mirror temporal changes or trends in surface-water chemistry reported by other studies from the region. Between 1992 and 2004, for example, concentrations of sulfate and nitrate from monthly samples decreased significantly in 100% and 56% of the 48 ALTM lakes, respectively, and corresponded to increased pH and ANC in 65% and 77% of the lakes, respectively (Driscoll et al.,

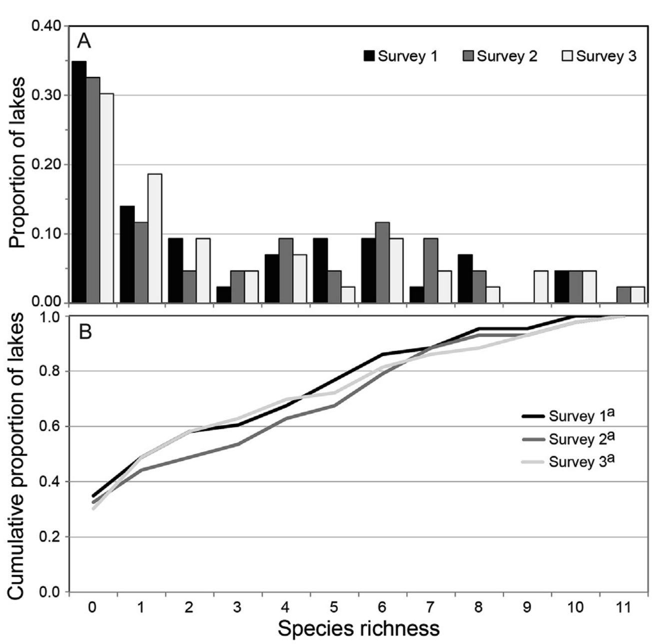

Fig. 6. The (A) proportion (frequency distribution) and (B) the cumulative proportion of 43 ALTM lakes with 0–11 fish species during three survey periods (1984–87, 1994–2005, and 2008–12). Different letters denote significant differences in the richness distributions among the three survey periods using two-sample Kolmogorov-Smirnov tests.

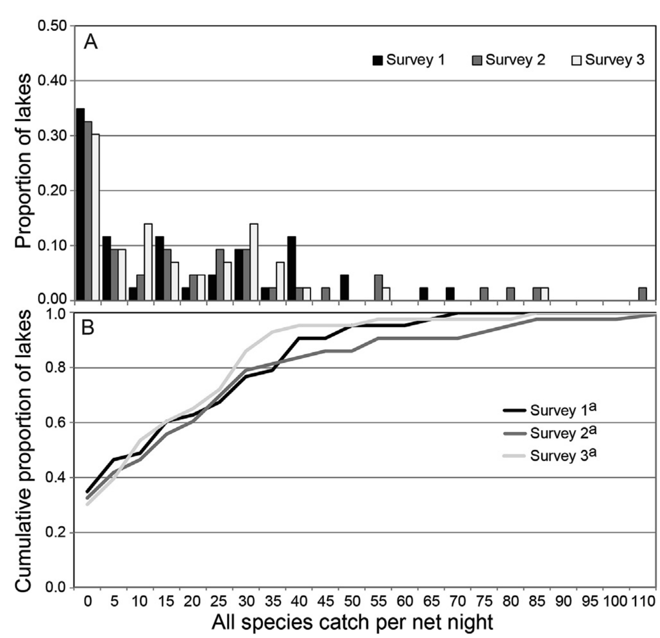

Fig. 7. The (A) proportion (frequency distribution) and (B) the cumulative proportion of 43 ALTM lakes with all species catch per net night (CPNN) of 0–110 fish during three survey periods (1984–87, 1994–2005, and 2008–12). Different letters denote significant differences in all species CPNN distributions among the three survey periods using two-sample Kolmogorov-Smirnov tests.

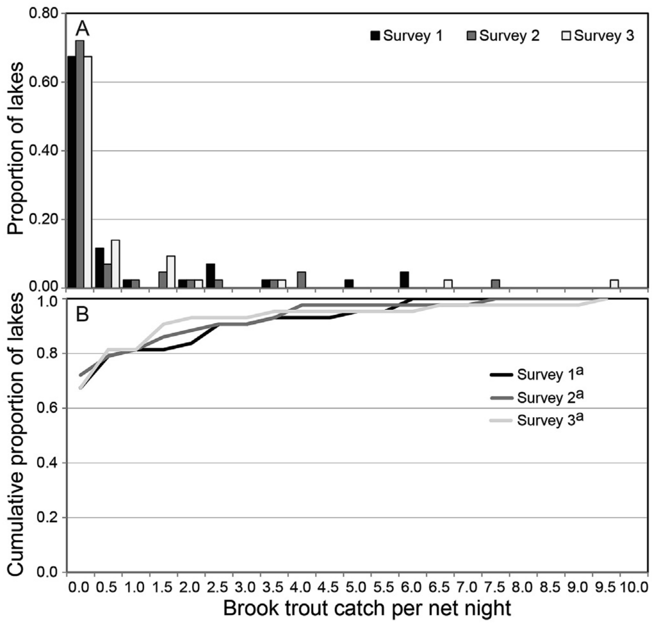

Fig. 8. The (A) proportion (frequency distribution) and (B) the cumulative proportion of ALTM lakes with all species catch per net night (CPNN) of 0–10 brook trout during the three survey periods (1984–87, 1994–2005, and 2008–12). Different letters denote significant differences in brook trout CPNN distributions among the three survey periods using two-sample Kolmogorov-Smirnov tests.

2007). The annual rate of decline in sulfate concentrations over the 22-year period (mean 2.16 μeq/L-y, or a total of about 26 μeq/L) was comparable to the rate of decline for 16 of the original 17 ALTM lakes (mean 2.02 μeq/L-y, or a total of about 44 μeq/L) between 1982 and 2004 and corresponded to increases in ANC of roughly 1.0 μeq/L-y for both periods (Driscoll et al., 2007). Similarly, Lawrence et al. (2011), reported that the mean ANC from 12 streams in the western Adirondacks, sampled 2–3 times per year during 1980–85 and again during 2003–05, increased on average by 13 μeq/L. The approximate rate of change in stream ANC concentrations (+0.57 μeq/L-y) was roughly half the rate of change in ANC for 48 ALTM lakes between 1992 and 2004 (+1.13 μeq/L-y) and between 1992 and 2013 (+0.95 μeq/L-y) (Driscoll et al., *in review*), but was more similar to the rate of change in ANC (+0.76 μeq/L-y) for another subset of 43 ALTM lakes between 1991 and 2007 that were widely distributed across the Adirondack Park (Waller et al., 2012). More important to fish communities were the significant decreases in Al_i concentrations (up to −0.20 μmol/L-y) in most of the 48 ALTM lakes (especially the 26 thin till drainage lakes) and indications that DOC concentrations were increasing in many of the ALTM lakes between 1992 and 2004 (Driscoll et al., 2007) and between 1992 and 2013 (Driscoll et al., *in review*). While increased supply of DOC and associated strongly acidic organic acids partially offset pH increases, higher DOC levels also increased organic complexation of Al and effectively decreaseed Al_i concentrations (Lawrence et al., 2007). The mean Al_i concentrations in 42 ALTM lakes declined from a range of 3–4 μmol/L during 1994–98 to less than 1.0 μmol/L during 2010–11, likely because hydrolysis and increases in DOC concentrations (in 74% of the lakes) decreased the Al_i fraction of total monomeric Al from 57% in 1994 to 23% in 2011 (Lawrence et al., 2013). The low Al_i levels noted by Lawrence et al. (2013) in 2010–11 were comparable to the mean summertime estimate of 0.66 μmol/L from the third (2008–12) survey in most of the same ALTM lakes. Because present day (2012) Al_i concentrations in many ALTM lakes average less than the commonly accepted threshold for brook trout survival (and for biological recovery in streams and lakes of the northeastern U.S.) of about 2.0 μmol/L (Baldigo et al., 2007; Driscoll et al., 2001), the water quality of many lakes may now be suitable for survival of some eradicated fish species and the recovery of their populations.

Fish communities and brook trout populations in the 43 ALTM lakes generally did not respond as hypothesized to improvements in water quality over the past 28 years. Analyses demonstrated that no fish metric changed significantly (positively or negatively); community richness averaged about 3 species, all species CPNN ranged from 14.5 to 22.0 fish per net night, and brook trout CPNN ranged from 0.60 to 0.69 fish per net night during each of the three surveys. Though changes in fishery metrics did not mimic shifts in acid-base chemistry, several positive trends were identified. For example, species richness increased in 33% of the study lakes and the percentage of lakes with no fish species declined from 35 to 30%, between the first and third surveys. In addition, all species CPNN increased in 30% of study lakes and brook trout CPNN increased in 19% of study lakes between the first and third surveys. The lack of significant changes in fish metrics from ALTM lakes was not entirely unexpected. Recent investigations rarely detected significant trends or changes in biological metrics in response to the chemical recovery of acidified surface waters. For instance, brook trout mortality during *in-situ* exposures to stream waters in the western Adirondacks was high and variable during 2001–03 and did not differ from toxicity tests conducted in the same streams during 1984–85, 1988–90, and 1997 (Baldigo et al., 2007). Sediment-core data from a 2007 paleolimnological study in one large Adirondack lake (ALTM lake 040752; Big Moose Lake, not included in our analysis) that is undergoing chemical recovery from acidic deposition indicates that chrysophyte and diatom assemblages are starting to recover, but that these two groups (and cladocerans) have not returned to their pre-acidification state (Arseneau et al., 2011). Sutherland et al. (2015) argued that ALTM lake 040874 (Brook Trout Lake, included in our analysis), which has recovered chemically and was uninhabited by fish for at least 20 years (1984–2004), could not have reestablished a brook trout population without stocking because it has poor connectivity to waters with source populations.

The lack of preacidification records was identified as a major obstacle to assessments of biological recovery in lakes across Sweden (Holmgren, 2014; Valinia et al., 2014). For example, Valinia et al. (2014) used historical fish records and hydrogeochemical models to show that biological recovery generally lagged behind chemical recovery. These researchers found that acid-sensitive roach (*Rutilus rutilus*) populations, absent from 14 of 28 lakes classified as acidified in 1980, became reestablished in only 5 of the 14 lakes that experienced chemical recovery by 2010. Holmgren (2014) concluded that while the chemistry of many lakes in Sweden have recovered from acidification, changing climate conditions were beginning to confound biological recovery. Analysis of data from three lakes in Sweden impacted by acidic deposition indicate that (a) pH has increased significantly (but it has not yet reached preindustrial levels); (b) temporal trends in fish and phytoplankton richness were not significant; (c) roach were lost from one of the three lakes; and (d) there were few direct relations between biological and chemical metrics between 1994 and 2013 (Holmgren, 2014). These investigators offered several plausible explanations as to why biological recovery has not occurred, or why recovery has been difficult to quantify in their study areas, which may also apply to Adirondack lakes.

There are a number of likely explanations for our inability to detect significant changes in fish communities or brook trout populations in ALTM lakes that have experienced moderate levels of chemical recovery. These reasons generally fall into two categories based on assumptions of whether or not biological recovery

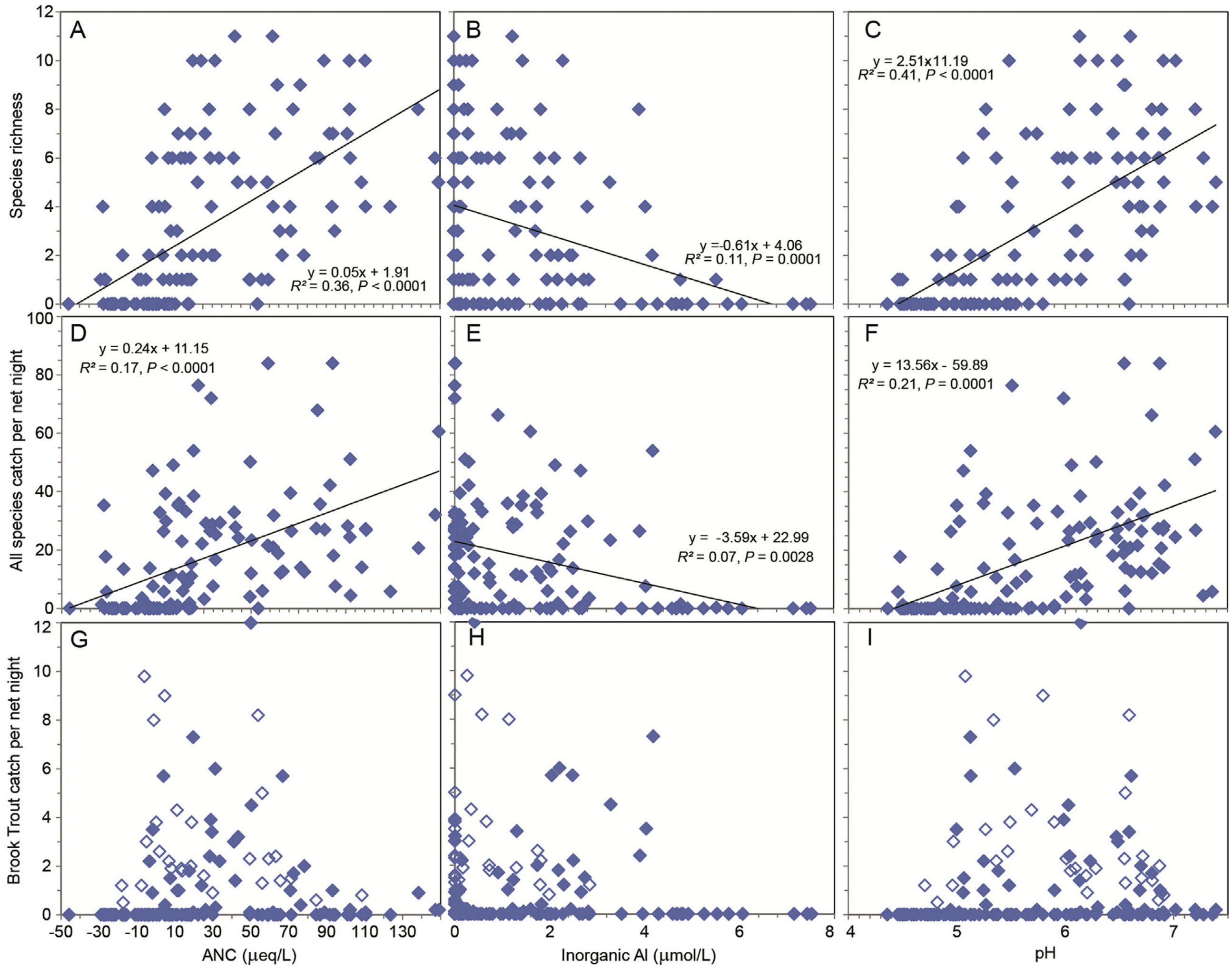

Fig. 9. The relations between total species richness and (A) acid neutralizing capacity (ANC), (B) inorganic Al (Al_i), and (C) pH; between all species catch per net night (CPNN) and (D) ANC, (E) Al_i, and (F) pH; and between brook trout CPNN and (G) ANC, (H) Al_i, and (I) pH from surveys in 43 ALTM during the three periods (1984–87, 1994–2005, and 2008–12). Brook trout data from stocked lakes were excluded from analyses and denoted by open diamonds in panels G, H and I.

has occurred. If biological recovery has occurred, our analyses may have been unable to identify significant temporal changes in target metrics because the fish-sampling design lacked sufficient statistical power to detect changes in these highly variable systems. Measures of sampling (for each survey) and temporal (among years) variability or for key fish metrics within individual lakes are currently unknown and cannot be estimated without additional (multiple) surveys over successive years or replicated surveys during a single year in representative lakes. Alternatively, more labor intensive mark-and-recapture methods, which typically target single species (Zale et al., 2013), might be employed to quantify density and biomass (and measures of error) for resident species populations and for entire lake communities.

The most logical explanation for our findings, however, is that fish assemblages have actually not recovered from acidification on a broad (regional) scale in Adirondack lakes. Many lakes of the region occur in high-elevation headwaters with few or no tributaries and numerous physical barriers in downstream (outlet) streams. Even after chemical recovery, many native species in such systems may be unable to navigate back to lakes from which they were extirpated and reestablish reproducing populations without human stocking efforts (Sutherland et al., 2015). Even though acid-base chemistry has improved substantially in the ALTM (and other) lakes, there could be a significant time lag for species populations, communities, and entire ecosystems to recover in disturbed systems once stressors have been eliminated (Yan et al., 2003). Barnthouse (2004), for instance, used the generation time approximation method to predict that populations of fish species with short (0.5 y) to long (5 y) generation times (1- to 7-y life spans) could theoretically recover to pre-disturbance levels after stressors were alleviated in about 2–11 y, respectively. The actual times for full recovery of populations of fish species in natural systems, however, were found to range from less than 1 year to more than 50 years (Niemi et al., 1990), and may reflect variations in the fraction of the species' population lost, whether or not the species was completely extirpated, differences in the maximum population growth rates (generation times) of colonist species, the mobility and size of colonist species, the proximity of source populations, interactions with established generalist taxa that are acid-tolerant, and the presence or absence of dispersal barriers (Barnthouse, 2004; Keller et al., 1998; Lundberg et al., 2000; Niemi et al., 1990; Yan et al., 2003). Accordingly, the recovery of brook trout, which may live for 4–9 years in Adirondack lakes, may be expected to take about 11 years after highly acidic and toxic Al_i conditions are eliminated. On the other hand, some minnow species with short life spans might be expected to reestablish populations within 2–3 years. An extensive analysis of monthly chemistry data from 43 ALTM lakes between 1994 and 2011 determined that mean Al_i concentrations first decreased below 2.0 μmol/L in 2007, and below 1.0 μmol/L in 2010 (Lawrence et al., 2013). Even if all other conditions were ideal (and acidic episodes did not occur), the time between the occurrence of survivable water-quality conditions (2007–10) and the third ALTM-lake survey (2008–12) is insufficient to permit the populations of many native fish species to fully recover. The period 2018–21 is the earliest that widespread recovery of long-lived species populations, such as acid-tolerant brook trout, might be anticipated, and 2012–13 may have been the earliest that broader recovery of short-lived and acid-intolerant fish species may have occurred. The actual recovery times for fish assemblages in ALTM lakes, however, would obviously vary due to the magnitude of chemical recovery, the effects of limiting factors (as noted above), and other changes in biological conditions (e.g., establishment of non-native species populations) within each lake since native species were eliminated. Potentially more important may be the inverse relation between brook trout CPNN and ANC or Al_i (Fig. 9G, H), which suggests that increases in ANC and decreases in toxicity might not benefit their populations in recovering lakes. Although speculative, improvements in water chemistry (decreases in toxicity) could promote rapid colonization of other less acid-tolerant generalist (native or non-native) species, which may be able to outcompete brook trout or possibly prey on their eggs or juveniles and effectually limit population densities in recovering lakes below their historic norms. Broad dispersion of non-native species across the Adirondacks (Daniels et al., 2008) and evidence that reproducing populations of brook trout may be hindered by changing thermal regimes in lakes (Robinson et al., 2010; Warren et al., 2012) add credence to the notion that the return of native species to recovering aquatic ecosystems (Holmgren, 2014) within the Adirondack region will be a highly complex phenomena.

The lack of measureable recovery of fish assemblages in the 43 ALTM lakes between 1984 and 2012 has important implications for lake and fisheries management and related assessment programs. First, the lack of observable biological recovery does not imply that the CAA and subsequent rules have not had the intended effects on water quality in Adirondack lakes. Although a broad recovery in lake fisheries was not apparent in our analyses, the water quality in most lakes has improved substantially over the past 2–3 decades (Driscoll et al., this issue; Waller et al., 2012) and created conditions that should help shepherd the restoration of pre-acidification ecosystems in lakes across the region. If changes in acid-base chemistry of some ALTM lakes were, in fact, too limited to have already affected resident biota, then additional management actions to accelerate chemical recovery (e.g., liming of lakes and watersheds or additional reductions in sulfur and nitrogen oxide emissions) might be warranted. Second, our findings suggest that the recovery of native fish species populations and functionally diverse food webs may require supplementary efforts to reintroduce native species, especially fish, which are easily blocked by barriers in headwater lakes with low connectivity (Gunn and Mills, 1998). Such a solution (stocking of mature brook trout) helped expand and diversify the abbreviated food web in Brook Trout Lake (ALTM lake 040874) as noted above (Sutherland et al., 2015). In contrast, brook trout populations in another completely isolated (headwater) Adirondack lake have recovered markedly because remnant populations in several tributaries with high ANC (refuges) provided a source of fish to reestablish their lake populations when acidity and Al_i levels decreased to survivable levels during the past decade (Josephson et al., 2014). Third, the lack of detectable biological recovery points out the need to formulate more defensible pre-acidification target conditions for judging progress towards (and barriers to) biological recovery in individual lakes or groups of lakes. Holmgren (2014), for example, suggested progress should be gauged based upon the pre-acidification abundance, recruitment and/or growth of “locally dominating” species. Fourth, the potential challenges to recovery of species' populations, such as: (a) inadequate water quality, (b) the presence of physical barriers that block colonists, (c) too few colonists to establish viable populations, or (d) community-level confounding factors, need to be identified and resolved within target lakes to ensure that meaningful biological recovery can proceed (Yan et al., 2003). Fifth, as noted in the earlier discussion, the current assessment strategy (i.e., sampling methods and frequency) cannot generate measures of error for key fishery metrics. This hinders our ability to accurately define current conditions and assess the significance of temporal changes in biological metrics within individual lakes and across groups of lakes. Future fish-survey methods and strategies, therefore, need to be critically reviewed and revised so that they will be able to generate accurate biological metrics with quantifiable levels of error.

Our findings have important ramifications for the derivation and interpretation of critical and target loads for deposition of nitrogen and sulfur to watersheds that are presently (2015–16) under review by the U.S. Environmental Protection Agency through the Clean Air Scientific Advisory Committee, Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur. Critical loads (CLs) could be used to establish secondary standards for S and N emissions, which will theoretically protect terrestrial and aquatic species (and their communities) from further adverse impacts and promote recovery of acidified ecosystems to an un-impaired or minimally acceptable condition. The standards, if implemented, will rely heavily on research which estimates thresholds or target deposition loads of nitrogen and sulfur, below which significant harmful effects on sensitive elements of terrestrial and (or) aquatic ecosystems should not occur in receiving watersheds. A number of acid-base chemistry parameters such as pH, ANC, and Al_i have corresponding effect thresholds for several fish species, which when exceeded are likely to impair their health, cause mortality, reduce recruitment rates and population density, shift species distributions, and decrease overall community diversity. The absence of notable recovery and high temporal variability in biological metrics from ALTM lakes, however, suggest that tangible biological responses to sulfur and nitrogen deposition loads that decrease below safe (threshold) levels may not be evident or detectable for years or decades after chemical recovery occurs in the region. The lack of rapid biological responses to improving chemistry in ALTM lakes suggests that one or more acid-base chemistry variables (noted above) may serve as acceptable surrogates for gauging the “potential” for biological responses to various target loads of sulfur and nitrogen deposition. Accordingly, the specific biological response variables, and the sampling methods and frequency used to assess such responses, need to be revised to more precisely characterize biological recovery from acidification in ALTM lakes, and in other lakes of the region.

The ability to accurately characterize and detect changes in highly variable fishery metrics such as species richness and abundance using gear that is selective to specific habitats or sizes (and age classes) of fish in lakes provokes the question: could biological recovery be more effectively quantified or detected in streams of the region? Temperate lakes have been the focus of long-term monitoring of acid-base chemistry partly because summer conditions are stable for relatively long periods. The lack of variability in lake hydrology means that temporal changes and trends in chemistry may be detected more easily in lakes than in streams. The relatively large volume and diverse/stratified habitats of lakes, however, make most biological-community surveys difficult and associated results imprecise. Most small fish species and early life stages of large species are seldom sampled effectively in lakes, whereas all large and small individuals can be efficiently collected in blocked stream reaches. Small fish species and early life stages of many fish are also often more sensitive to acidic conditions than are larger species and later life stages (Baker et al., 1996; Baldigo and Lawrence, 2001). In the United Kingdom, Malcolm et al. (2014), showed that brown trout fry were more sensitive indicators of biological recovery than parr, and Monteith et al. (2005) found that juvenile brown trout were the earliest immigrants, in 3 of 22 streams (and lakes outflows) beginning to recover from acidification. Juveniles of the native and widely distributed brook trout have been used for more than three decades to quantify toxicity and biological impacts of acidification in Adirondack streams (Baker et al., 1996; Baldigo et al., 2007; Johnson et al., 1987), thus, it would make an ideal indicator species for the region. Although stream chemistry is more dynamic temporally and less responsive than lake chemistry to decreasing trends in acidic deposition, stream habitat is less dimensional than lakes, thus, most life stages, species populations, and biological communities (especially fish) are much easier to quantify in streams than in lakes. Monitoring and analysis of temporal trends in stream biota, thus, could improve our ability to detect and characterize indicators of biological recovery in surface waters of the Adirondack region that are recovering from acidification.

In summary, the CAA and other federal regulations have clearly reduced sulfur and nitrogen oxide emissions, acidic deposition, and the acidity and toxicity of waters in the ALTM lakes, but these changes have not triggered widespread recovery of brook trout populations or fish communities. The lack of detectable biological recovery appears to result from relatively recent chemical recovery and an insufficient period for species populations to take advantage of now-suitable water quality. Recovery of extirpated species' populations may simply require more time for individuals to immigrate to and repopulate formerly occupied lakes. Supplemental stocking of selected species may be required in some lakes with no remnant (or nearby) populations or with physical barriers between the recovered lake and source populations. The lack of detectable biological recovery could also be related to our inability to calculate measures of uncertainty or error and, thus, examine temporal changes or differences in populations and community metrics in more depth (e.g., within individual lakes) using existing datasets. Indeed, recovery of several brook trout populations and partial recovery of fish communities have been documented in several lakes of the region, both with and without human intervention. Multiple fishery surveys or the use of mark and recapture methods within individual lakes would help alleviate the issue (provide measures of error for key fishery metrics) within the context of a more focused sampling strategy. Although stream chemistry is more dynamic temporally and less responsive than lake chemistry to decreasing trends in acidic deposition, stream habitat is less dimensional than lakes, thus, most life stages, species' populations, and biological communities (especially fish) are relatively easy to quantify. Such long-term monitoring efforts could increase our ability to detect and quantify biological recovery in neutralizing surface waters throughout the Adirondack Region.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2016.06.049.

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