

# Glasgow Lake: an early-warning sentinel of lake acidification in Cape Breton Highlands National Park (Nova Scotia, Canada)

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**Abstract** In contrast to other lakes studied in Cape Breton Highlands National Park (Nova Scotia, Canada), our paleolimnological results indicated that Glasgow Lake has been impacted by acidic deposition starting in the early 1900s. Based on analysis of diatom assemblages, the lake experienced a decrease in diatom-inferred lakewater pH from a pre-industrial pH of  $\sim 5.8$  to a current pH of 5.3 (2000–2002 measured mean pH = 5.0) as well as a decrease in diatom-inferred Gran-alkalinity. In this study, diatom-based paleolimnological techniques were used in conjunction with a dynamic biogeochemical model (MAGIC) to assess both the timing and extent of the acidification trend, as well as determine a probable explanation as to why this lake, and none of the other 15 Cape Breton Highlands lakes studied for

paleolimnology thus far, acidified under a peak non-marine sulphate deposition load of  $43.6 \text{ mmol}_c \text{ m}^{-2} \text{ year}^{-1}$  in the mid-1970s. Steady-state models estimate that Glasgow Lake had the lowest buffering capacity of six study lakes and estimated critical sulphate loading of  $<1 \text{ mmol}_c \text{ m}^{-2} \text{ year}^{-1}$ . MAGIC also estimated a loss of charge balance alkalinity from a pre-1850 value of  $38 \text{ } \mu\text{mol}_c \text{ l}^{-1}$  to a low of  $12 \text{ } \mu\text{mol}_c \text{ l}^{-1}$ . While no evidence of biological recovery has been recorded, MAGIC estimates an increase in charge balance alkalinity to  $27 \text{ } \mu\text{mol}_c \text{ l}^{-1}$  in 2002 in response to decreased  $\text{SO}_2$  emissions. Of the five other lakes that were modelled, all showed trends towards more acidic states and subsequent increases in charge balance alkalinity; however, the empirical paleo-diatom approach applied to these lakes showed no evidence of acidification. Thus, Glasgow Lake has the lowest buffering capacity among the Cape Breton Highland study lakes and serves as a sentinel of potential acidification trends and recovery in this region.

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## Introduction

Surface-water acidification due to deposition of anthropogenic-sourced strong acid anions has affected the ecology and chemistry of lakes in Nova Scotia, a

province in the Atlantic region of Canada. In this province, sulphate ( $\text{SO}_4^{2-}$ ) deposition is generally highest in the southwest [ $\sim 39.5 \text{ mmol}_e \text{ m}^{-2} \text{ year}^{-1}$  ( $19.0 \text{ kg ha}^{-1} \text{ year}^{-1}$ ) (2000–2002), with peak deposition estimated at  $63.6 \text{ mmol}_e \text{ m}^{-2} \text{ year}^{-1}$  ( $30.5 \text{ kg ha}^{-1} \text{ year}^{-1}$ ) in 1973], a region that is closest to major emission sources (in central Canada, midwestern USA and northeastern USA), and thus receives higher levels of  $\text{SO}_4^{2-}$  deposition introduced by long-range atmospheric transport (Clair et al., 2001). Sulphate deposition decreases towards the northeast of the province with the exception of an area east and downwind of local  $\text{SO}_4^{2-}$  sources at the city of Halifax (Clair et al., 2001). Previous studies by Ginn et al. (2007a, b, c) and Tropea et al. (2007) identified major environmental changes due to acidic inputs in lakes in Kejimikujik National Park and around Halifax, but little evidence of widespread lake acidification in other areas of the province. In these studies, before-and-after paleolimnological techniques (i.e. top–bottom sediment analysis, see Smol, 2008 and Cumming et al., 1992) were used to compare current biological assemblages of diatoms and associated inferences of pH with pre-industrial (i.e. pre-1850) assemblages (Ginn et al., 2007a). Of the 16 paleolimnology study lakes in Cape Breton Highlands National Park, Glasgow Lake was the only study site that recorded evidence of diatom-inferred acidification since the 1800s (Ginn et al., 2007a). In addition to the 16-site before-and-after paleolimnological studies, six of these lakes were used for a decadal-scale detailed analysis, although changes in the diatom assemblages of these lakes were related to climatic influences rather than surface-water acidification (Ginn et al., 2007b). Paleolimnological techniques have not only been used to assess impacts from acidification, but also a wide variety of environmental issues such as drainage from mine sites (Salonen et al., 2006), increased nutrient loading (e.g. Miettinen et al., 2005), climatic changes (e.g. Laird et al., 2003; Moos et al., 2005; Smol & Douglas, 2007), fish populations (e.g. Gregory-Eaves et al., 2003), organic pollutants (e.g. Krümmel et al., 2005) and other lake management issues (e.g. Ginn et al., 2008).

In addition to the paleolimnological analyses, biogeochemical modelling techniques were used to assess the impact of acidic deposition across Nova Scotia, including six lakes in Cape Breton Highlands National Park. Whitfield et al. (2006a) presented

critical loadings of  $\text{SO}_4^{2-}$  for the region where Glasgow Lake was demonstrated to have the lowest critical load among the six modelling study lakes. Additionally, the dynamic Model of Acidification of Groundwater In Catchments (MAGIC: Cosby et al., 1985) was used to reconstruct pre-acidification chemical conditions of the lakes, as well as to estimate temporal changes during the period 1850–2100 (Whitfield et al., 2007).

The objectives of this study were to examine the timing and nature of acidification trends recorded in Glasgow Lake, which had previously only been inferred based on an estimate of pre-industrial and present-day pH inferences from diatom assemblages (Ginn et al., 2007a), and to determine why this lake, and no other study lake in Cape Breton Highlands National Park, recorded a decrease in diatom-inferred pH.

## Material and methods

### Study site and sediment core collection

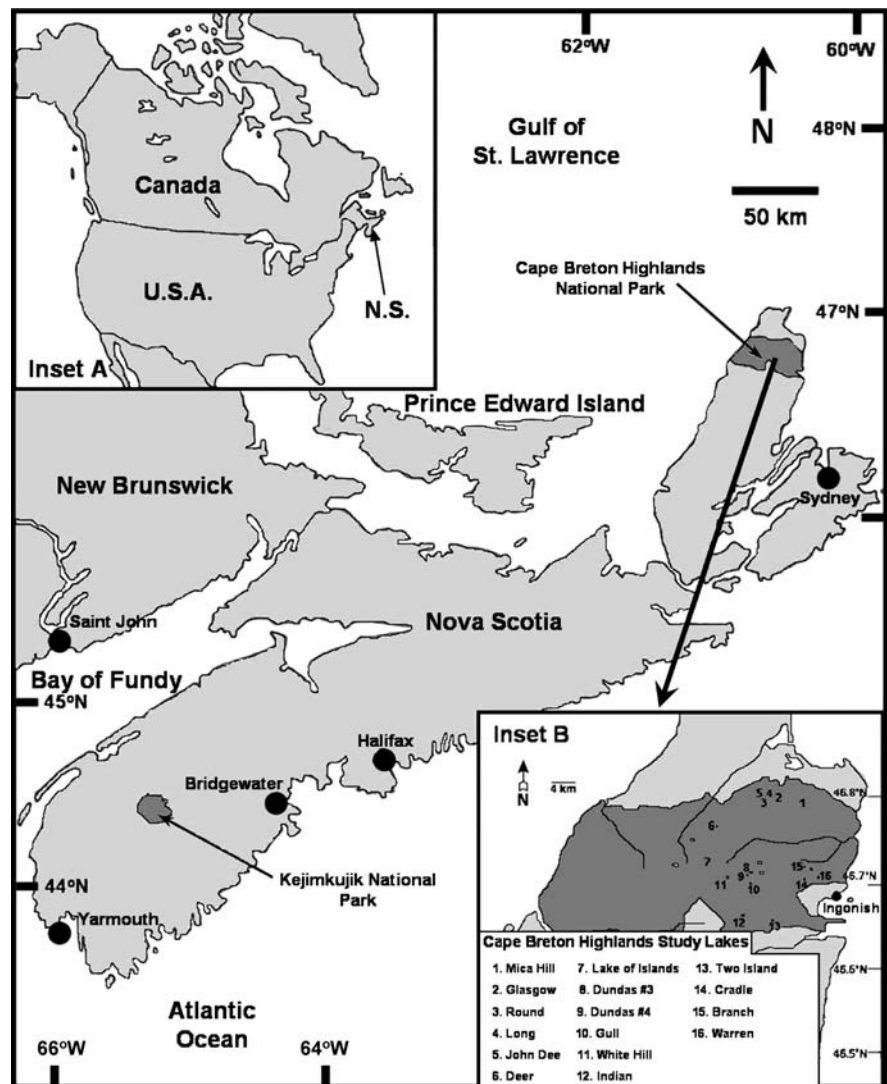
Glasgow Lake ( $46^\circ 49.2' \text{ N}$ ,  $60^\circ 29.5' \text{ W}$ , elevation: 419 m above sea level) is located on the taiga plateau of Cape Breton Highlands National Park (Fig. 1; Table 1). Mean long-term precipitation and runoff are 1.44 and 1.06 m  $\text{year}^{-1}$ , respectively. While this park has a variety of ecosystems ranging from coastal forests and seashores, the taiga plateau has elevations 300–500 m above sea level and a climate more typical of subarctic regions with long winters, high winds and a sudden spring freshet in late April or May. Vegetation in this region consists of shrubby conifers, *Sphagnum* bogs and subarctic scrub (Davis & Browne, 1996).

The sediment core from Glasgow Lake was collected through 1.5 m of ice in March 2004 using a Glew gravity corer (Glew, 1989; Glew et al., 2001) equipped with a 7.6-cm internal diameter, 50-cm long Lexan<sup>®</sup> tube. The core was sectioned at 0.25 cm intervals using a Glew (1988) extruder with separate intervals placed in individual Whirl-Pak<sup>®</sup> bags, stored on ice and kept  $\sim 4^\circ \text{C}$  for transport back to the lab and subsequent storage.

### Radiometric dating

Sediment samples were dried using a Virtus<sup>®</sup> freeze dryer, and  $\sim 0.5 \text{ g}$  of each core interval was placed in

**Fig. 1** Map showing the location of Cape Breton Highlands National Park and other study regions, in Nova Scotia, Canada. Inset A shows the location of Nova Scotia (NS) within the context of North America. Inset B shows the location of the 16 study lakes in Cape Breton Highlands National Park



plastic test tubes for  $^{210}\text{Pb}$  gamma dating following procedures outlined by Schelske et al. (1994). Activities of  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$ ,  $^{214}\text{Bi}$  and  $^{137}\text{Cs}$  were calculated based on procedures outlined by Schelske et al. (1994) using an Ortec<sup>®</sup> germanium (Gr) crystal well detector for 80,000 s. Dates were calculated using the constant rate of supply (CRS) model with a computer program developed by Binford (1990, University of Florida).

#### Diatom preparation and analysis

Diatoms were isolated from the sediment using a 1:1 molar ratio of  $\text{HNO}_3$  and  $\text{H}_2\text{SO}_4$  as described by Battarbee et al. (2001) and mounted on slides using

Naphrax<sup>®</sup>. A minimum of 300 diatom valves were enumerated per sample using a  $100\times$  oil immersion objective ( $\text{NA} = 1.3$ ) on a Leica<sup>®</sup> DMRB microscope equipped with DIC optics. Diatoms were identified to the lowest taxonomic level possible using Patrick & Reimer (1966, 1975), Krammer & Lange-Bertalot (1991–2000), Round et al. (1990), Camburn & Charles (2000), Fallu et al. (2000), Wehr & Sheath (2002) and other references fully described in Ginn et al. (2007c).

#### Statistical analysis and inference of pH and Gran-alkalinity

Lakewater pH was identified in a canonical correspondence analysis (CCA) as the most important

limnological variable influencing the distribution of diatom taxa in a dataset from northeastern North America, a dataset used to develop diatom-based pH (and other limnological variables) inference models based on a number of approaches (the NENA dataset; Ginn et al., 2007c). Here, we use the maximum likelihood model since it was the most robust model, although other techniques yielded similar results (Ginn, 2006; Ginn et al., 2007c). This pH model has strong predictive ability ( $r^2_{\text{boot}} = 0.88$ ; RMSEP = 0.45) and was highly significant ( $P < 0.001$ ). The Gran-alkalinity model was slightly less robust ( $r^2_{\text{boot}} = 0.75$ ; RMSEP = 0.38) but still significant ( $P < 0.05$ ). Relative frequency diagrams of the dominant diatom taxa (>5%) were constructed using the computer program C2 (version 1.4; S. Juggins, University of Newcastle), with diatoms arranged by principal components analysis (PCA) axis-1 species scores. PCA axis-1 site scores were used to track downcore variation. PCA of the 11 measured chemical and physical limnological characteristics was also used to assess if these characteristics differed between Glasgow Lake and the other 15 study lakes from Cape Breton Highlands National Park.

### Biogeochemical modelling

Biogeochemical models were applied to a subset of six of the Cape Breton lakes investigated through paleolimnological analyses. Critical loads for sulphur

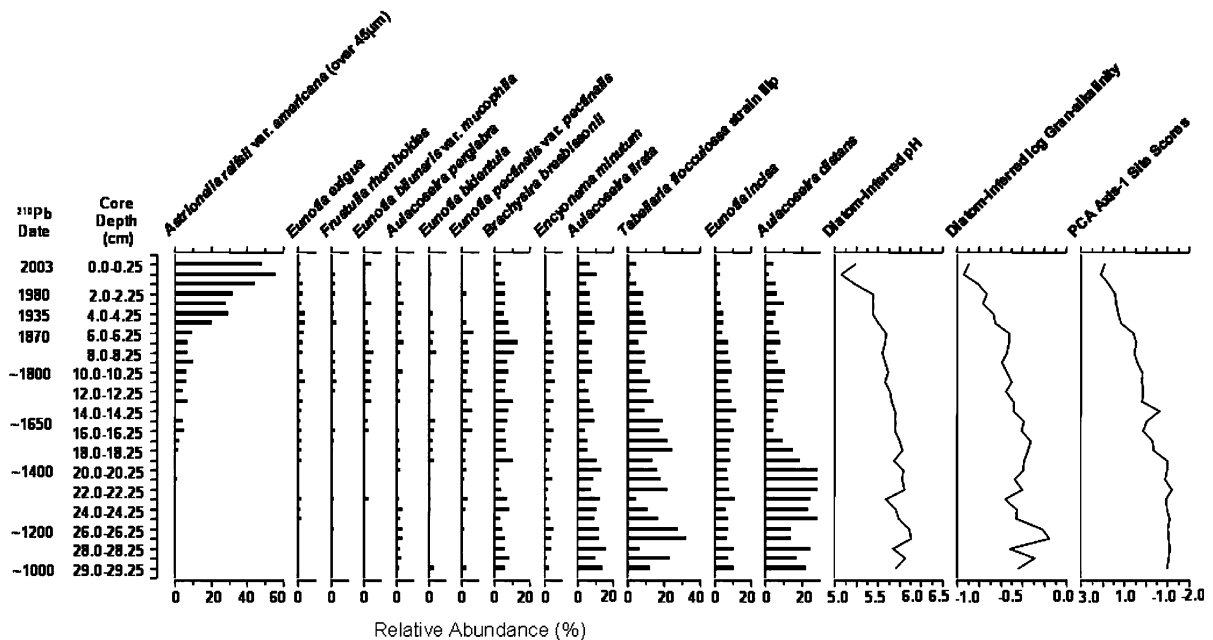
(S) were calculated using the steady-state water chemistry model (SSWC; Henriksen & Posch, 2001), which is largely empirical, and bases estimates of the critical load of S on the current base cation flux from the catchment. Five-year (1998–2002) mean annual lake chemistry and long-term average runoff volume were used as model inputs. Full details are provided in Whitfield et al. (2006a). MAGIC was used to predict historical and future chemical conditions of the lakes and calibrated to a long-term time series of observed lake chemistry (1990–2002) and catchment soil base saturation (2004). See Whitfield et al. (2007) for a complete description of the modelling procedure. It is important to note that MAGIC estimates the acid neutralizing capacity, referred to here as the charge balance alkalinity ( $\text{Alk}_{\text{CB}}$ ) and calculated as the difference between the sums of base cation charges and acid anion charges, while the paleolimnological assessment estimates Gran alkalinity (Alk). Alk and  $\text{Alk}_{\text{CB}}$  generally agree well except for low acid neutralizing capacity waters, as measurements of Alk do not include the influence of dissolved organic carbon or aluminium (Sullivan et al., 1989) and Alk may overestimate the buffering capacity of the water (Neal et al., 1999). Thus, these two measures are not directly comparable.

### Results

A total of 82 diatom taxa were recorded in the sediment core from Glasgow Lake, most of which were rare. Some modest changes in diatom assemblages occurred in the pre-industrial period (i.e. changes in dominance between *Aulacoseira distans* and *Tabellaria flocculosa* strain IIIp between ~1200 AD and ~1400 AD); however, these did not result in a large change in diatom inferred pH (Fig. 2). The more recent pre-industrial (~1800–1850) diatom assemblage was dominated by *A. distans* and *T. flocculosa* strain IIIp which started changing to an increased abundance in *Asterionella ralfsii* var. *americana* (>45  $\mu\text{m}$ ) at a depth of ~5 cm, corresponding to the early 1900s (Fig. 2). Further increases in the relative abundance of this taxon occurred in ~1960. Corresponding to this change in diatom assemblage, diatom-inferred lake-water pH decreased from a pre-industrial pH = 5.8 to a present-day pH = 5.3, which is close to the current (2000–2002) measured lakewater pH of 5.0. The PCA

**Table 1** Physical and limnological data from Glasgow Lake, Cape Breton Highlands National Park, Nova Scotia, Canada (water chemistry values are 3-year (2000–2002) annual averages)

Variable	
Mean annual precipitation	1.44 m year <sup>-1</sup>
Long-term annual runoff	1.06 m year <sup>-1</sup>
Mean depth	4.5 m
Surface area	23.9 ha
Watershed area	143 ha
Water residence time	1.43 year
Elevation (above sea level)	418 m
Total phosphorus (TP)	6.5 $\mu\text{g l}^{-1}$
pH	5.0
Dissolved organic carbon	6.5 mg l <sup>-1</sup>
Specific conductivity	25.1 $\mu\text{S cm}^{-1}$
Gran alkalinity	-12 $\mu\text{mol}_e \text{l}^{-1}$



**Fig. 2** Relative abundances of the dominant diatom taxa (>5%) in the sediment core from Glasgow Lake, Nova Scotia, Canada. Also included are diatom-inferred pH, diatom-inferred

log Gran-alkalinity and PCA axis-1 site scores, which show the main direction of variation

axis-1 site scores summarize the main direction of variation in the downcore diatom assemblages and has a highly significant correlation ( $r = 0.9$ ,  $P < 0.001$ ) to the pH inferences (Fig. 2). Results of PCA analysis on the physicochemical variables of all 16 study lakes revealed Glasgow Lake separating from other lakes based on a small watershed area, large volume, low catchment area to lake area ratio and high retention time (or slow flushing rate) (Fig. 3).

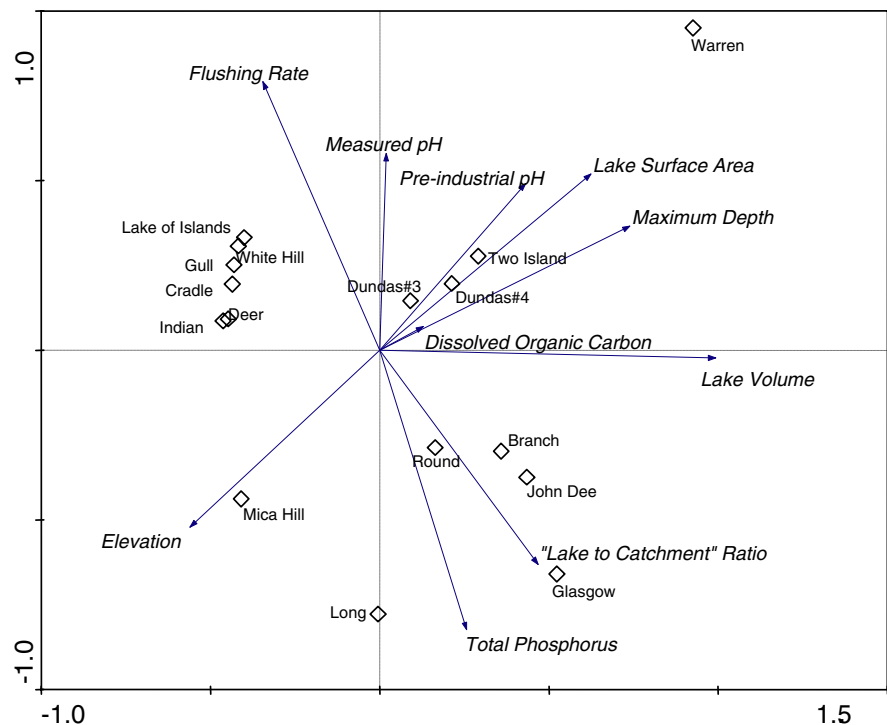
When differences in Alk were examined between the 16 Cape Breton study lakes (Ginn, 2006), Glasgow Lake had the lowest pre-industrial diatom-inferred Alk as well as the lowest current diatom-inferred Alk (Table 2). Geochemical modelling supports these findings; pre-industrial  $\text{Alk}_{\text{CB}}$  was estimated to be lowest at Glasgow Lake for all the Cape Breton Highlands study lakes. Further, the estimated critical  $\text{SO}_4^{2-}$  load (SSWC) for Glasgow Lake was the lowest of all study lakes in Cape Breton Highlands National Park (Table 2). Using MAGIC, all Cape Breton lakes show a loss in  $\text{Alk}_{\text{CB}}$  due to acidic inputs, and five of six lakes show noticeable decline in pH (Whitfield et al., 2007). The model hindcast shows evidence of decreases in both  $\text{Alk}_{\text{CB}}$  and pH at Glasgow Lake through the mid-1970s

(Fig. 4), coinciding with maximum sulphur emissions. At Glasgow Lake, MAGIC predicted a maximum pH depression of 0.5 units which is similar to the decrease inferred by paleolimnological techniques. Following the predicted bouts of acidification, MAGIC indicates subsequent increases in  $\text{Alk}_{\text{CB}}$  and pH, although both remain depressed in comparison to modelled historic levels.

## Discussion

In contrast to the other 15 lakes we studied in Cape Breton Highlands National Park, Glasgow Lake has a very low pre-industrial buffering capacity and, consequently, was very susceptible to even small loads of acidic deposition. The inability of this lake to neutralize acid inputs resulted in a diatom-inferred pH decrease of 0.5 units starting in the early 1900s and accelerating in the 1960s, a time of increased  $\text{SO}_4^{2-}$  deposition. While the  $\text{SO}_4^{2-}$  deposition in this area did not result in changes in the diatom assemblages of most lakes, unchecked increases in sulphur emissions may have soon overwhelmed the ability of other lakes to neutralize acidic inputs and resulted in

**Fig. 3** Principal components analysis (PCA) of 10 physical and chemical variables for the 16 study lakes from Cape Breton Highlands National Park

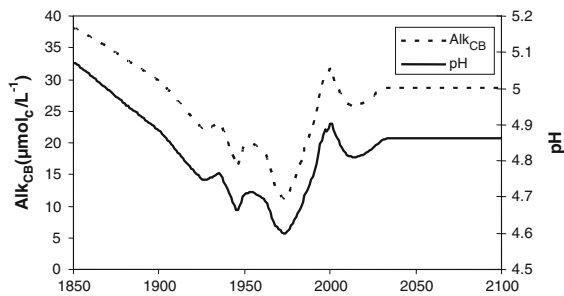


**Table 2** Three years (2000–2002) observed, pre-1850 and current diatom-inferred Gran alkalinity (Alk) for the 16 study lakes from Cape Breton Highlands National Park (Nova Scotia, Canada: Ginn, unpublished data), MAGIC estimated charge

balance alkalinity ( $\text{Alk}_{\text{CB}}$ ) (from Whitfield et al., 2007) and SSWC model estimated critical loads (CL) (from Whitfield et al., 2006a) for six of the study lakes

Lake	Mean Alk ( $\mu\text{mol}_c \text{ l}^{-1}$ )	Diatom inferred Alk		MAGIC estimated $\text{Alk}_{\text{CB}}$			SSWC CL ( $\text{mmol}_c$ $\text{m}^{-2} \text{ year}^{-1}$ )
		Pre-1850 ( $\mu\text{mol}_c \text{ l}^{-1}$ )	Current ( $\mu\text{mol}_c \text{ l}^{-1}$ )	1850 ( $\mu\text{mol}_c \text{ l}^{-1}$ )	1975 ( $\mu\text{mol}_c \text{ l}^{-1}$ )	2002 ( $\mu\text{mol}_c \text{ l}^{-1}$ )	
Branch	−24.2	7.6	5.2	56	28	44	5.3
Cradle	5.4	9.8	5.8				
Deer	49.4	10.4	10.2				
Dundas #3	−2.6	9.0	7.0				
Dundas #4	−2.0	7.8	6.6				
Glasgow	−12.2	5.2	1.8	38	12	27	0.4
Gull	−4.2	5.4	4.0				
Indian	−0.6	5.8	6.2				
John Dee	20.0	7.2	6.2	72	41	59	36.6
Lake of Islands	−14.2	8.2	7.6				
Long	66.6	7.0	6.8	144	102	119	74.5
Mica Hill	28.8	7.0	8.0	79	46	64	41.3
Round	−2.8	6.4	6.8	67	38	54	13.1
Two Island	−14.2	9.2	5.8				
Warren	33.2	42.4	14.2				
White Hill	−12.0	9.2	7.0				





**Fig. 4** MAGIC simulated charge balance alkalinity ( $\text{Alk}_{\text{CB}}$ ) and pH for Glasgow Lake, Cape Breton Highlands National Park, for the period 1850–2100

a widespread acidification trend in Cape Breton Highlands National Park. The implementation of  $\text{SO}_4^{2-}$  emission restrictions in the 1980s and 1990s likely averted this trend for the other Cape Breton lakes. MAGIC modelling has estimated a loss of  $\text{Alk}_{\text{CB}}$  since pre-industrial times but has suggested an increase in  $\text{Alk}_{\text{CB}}$  since 1975 (Fig. 4). Lake water chemistry monitoring began in 1990 at Glasgow Lake; thus, this limited data set would not have captured these changes. Additionally, trends in water chemistry are sometimes confounded by the influence of major sea-salt events (Evans et al., 2001), which may obscure any recovery in  $\text{Alk}_{\text{CB}}$  during the 1990s at Glasgow Lake (Whitfield et al., 2006a). While no biological recovery has been observed to coincide with the predicted chemical recovery from acidification at Glasgow Lake, biological recovery is expected to be delayed (Keller et al., 1999) and it may be many years before detection. The increase in Alk is also estimated in most other lakes which did not record a significant change in lakewater pH. In addition, all lakes show a decrease, albeit very small, in diatom-inferred Alk since 1850 (Table 2).

In Glasgow Lake, the change in species composition from *Aulacoseira distans* to *Asterionella ralfsii* var. *americana* (>45  $\mu\text{m}$ ) corresponds with an acidification trend and mirrors patterns observed in other acidified Nova Scotia lakes, which have high concentrations of dissolved organic carbon (DOC) (e.g. Pebbleloggitch, Peskowsk and Kejimikujik lakes in Kejimikujik National Park; Ginn et al., 2007a, c). While these lakes in Kejimikujik Park are naturally acidic due to the presence of organic acids in DOC (pre-1850 diatom-inferred lakewater pH  $\sim 5.8$ ), they have acidified further in response to  $\text{SO}_4^{2-}$  deposition. In fact, the minor increase in *Asterionella ralfsii* var.

*americana* (>45  $\mu\text{m}$ ) beginning at the 17-cm depth ( $\sim 1,500$  AD) may indicate a long-term trend in natural acidification. In some lakes, changes in the diatom assemblage suggest that the lake underwent a degradation of DOC (humic, coloured water to clearwater) in response to acidification (e.g. Pockw-ock Lake in Halifax; Tropea et al., 2007).

The timing of acidification in Glasgow Lake is estimated to be in the early 1900s, which would be expected from a very acid-sensitive lake (Fig. 2), and corresponds with increasing  $\text{SO}_4^{2-}$  deposition in Cape Breton (e.g. Whitfield et al., 2007). While Cape Breton Island receives lower  $\text{SO}_4^{2-}$  deposition [ $\sim 27 \text{ mmol}_\text{c} \text{ m}^{-2} \text{ year}^{-1}$  ( $13.0 \text{ kg ha}^{-1} \text{ year}^{-1}$ )] than the southwestern parts of Nova Scotia, Glasgow Lake is more acid-sensitive than other lakes in Cape Breton Highlands National Park with both the lowest pre-1850 diatom-inferred Alk and MAGIC estimated  $\text{Alk}_{\text{CB}}$  (Table 2). Predictions from MAGIC also indicate a very low pre-industrial pH ( $\sim 5.1$ ). The other 15 lakes studied in Cape Breton (Ginn, 2006) did not record a pH trend, although the diatom-inferred alkalinities show a small decrease in Alk in many lakes compared to pre-1850 values (Table 2). In contrast, lakes in the southwestern Nova Scotia (especially in Kejimikujik National Park), an area of relatively higher  $\text{SO}_4^{2-}$  deposition, have recorded a widespread acidification trend starting  $\sim 1925$ –1940 (Ginn et al. 2007b). The low buffering capacity of Glasgow Lake is due in part to granitic bedrock and shallow, base-poor soils in its catchment. The combination of the above factors yields relatively small base cation pools in the catchment soil, as is common across Nova Scotia where weathering rates are often very low (Whitfield et al. 2006b). As a result, the contribution of base cations to runoff is limited and the estimated critical load for the catchment is only  $0.4 \text{ mmol}_\text{c} \text{ m}^{-2} \text{ year}^{-1}$  (Table 2), among the lowest of the 20 catchments evaluated across the province. Thus, the physical characteristics of Glasgow Lake minimize the influence of the terrestrial drainage basin on water chemistry.

Glasgow Lake has one of the smallest catchment to lake ratios and the longest water residence time of the 16 study lakes (Fig. 3). The lake is much deeper than many of the other study lakes and, with its low flushing rate ( $0.70 \text{ year}^{-1}$ ), essentially acts as a bathtub, with much of the  $\text{SO}_4^{2-}$  deposited directly on the lake without being neutralized by interaction with the terrestrial catchment. While removal of  $\text{SO}_4^{2-}$  from lake water increases with

increasing water residency time, the rate of removal through in-lake alkalinity generation is still quite short compared to the range used by Kelly et al. (1987). Thus, with a large proportion of  $\text{SO}_4^{2-}$  deposited directly onto the lake surface and little ability to generate alkalinity via in-lake processes reducing sulphur,  $\text{SO}_4^{2-}$  likely becomes more concentrated in the lake during times of heavy loading, promoting decreases in alkalinity and pH.

MAGIC infers some degree of recovery from acidification for lakes in Cape Breton Highlands National Park (Table 2), while results from the paleolimnological analyses have not indicated increases in Alk, although depression during the period of elevated acidic deposition has been small; likewise, no evidence of biological recovery at Glasgow Lake has been recorded. Potential recovery of lakes in this region will be limited, as base cation release from the soil has decreased in response to decreased  $\text{SO}_4^{2-}$  deposition (Clair et al., 2001), and generation of base cations from bedrock and soil is minimal (Whitfield et al., 2006b). Whitfield et al. (2007) reported that recovery patterns of the lakes beyond emission reductions scheduled through 2030 are predicted to be small, and in the case of Glasgow Lake, no recovery is expected past 2030 without additional emissions reductions.

The acidification of Glasgow Lake in Cape Breton Highlands National Park can potentially serve as a warning of a possible regional acidification, if  $\text{SO}_4^{2-}$  deposition occurs at higher than present levels since lakes in this region are so poorly buffered. Long-term monitoring of lake chemistry and acidic deposition in this area will prove valuable in determining critical loadings to prevent future acidification of these lakes.

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