

Seventy-Four Years of Calcium Loss from Forest Soils of the Adirondack Mountains, New York

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In 2005/6, we conducted a remeasurement investigation of organic and mineral horizon Ca at 54 sites in forests of the Adirondack Mountains, New York. This network of plots represents northern hardwood, pine-dominated, and high-elevation spruce–fir stands typical of this region. Soil Ca had been measured at these sites twice (1930–1932 and 1984) in the previous 74 yr. Consistent with the original methods, we used measurements of dilute HCl (0.2 mol L^{-1}) extractable Ca to make comparisons among the 1930 to 1932, 1984, and 2005/6 samples. Between 1932 and 2005/6, median HCl-extractable Ca content decreased significantly (78%) in organic horizons and in the whole profile (organic + mineral horizons, 64%), although significant decreases in mineral soil Ca content were confined to the upper 20 cm. Moreover, significant decreases in organic horizon ($-1.5\% \text{ yr}^{-1}$), mineral horizon ($-1.0\% \text{ yr}^{-1}$), and whole profile ($-1.5\% \text{ yr}^{-1}$) Ca content between 1984 and 2005/6 indicated that Ca depletion has been sustained in recent decades. When stratified by forest type, there were significant decreases in extractable Ca concentrations and content in organic horizons and in the whole profile for all forest types between 1932 and 2005/6, although decreases between 1984 and 2005/6 were not necessarily significant. Overall, the rate of median whole-profile Ca depletion ($\sim 13.6 \text{ kg ha}^{-1} \text{ yr}^{-1}$) was similar during all sampling intervals, suggesting that the combined Ca inputs from atmospheric deposition and mineral weathering have not kept pace with leaching and sequestration in biomass during much of the 20th and early 21st centuries in these plots.

Abbreviations: HBEF, Hubbard Brook Experimental Forest; NC, Newcomb; NH, northern hardwood; OM, organic matter; PW, pine-dominated; SF, spruce–fir.

Calcium is an important nutrient for maintaining the vigor of some tree species and it plays an important role in buffering the effects of acidic deposition (Johnson et al., 1991), both of which are important considerations in forests of the Adirondack Mountains. Numerous investigations have identified the depletion of Ca and other base cation in forest soils of the northeastern United States as a threat to forest health through a number of mechanisms (Pitelka and Raynal, 1989; Johnson, 1992; Johnson and Lindberg, 1992; Eagar and Adams, 1992; Cronan and Grigal, 1995; Gbondo-Tugbawa and Driscoll, 2003; Tomlinson, 2003; Horsley et al., 2002; Drohan et al., 2002). Moreover, there is evidence that forest soil Ca depletion and acidification have continued in recent decades throughout the Adirondacks (Johnson et al., 2008a; Warby et al., 2009) despite reductions in atmospheric S emissions during the past three decades (Likens et al., 1996; Driscoll et al., 2001).

In northeastern U.S. forest soils, the supply of plant-available Ca and other base cations is primarily influenced by inputs from atmospheric deposition and mineral weathering. Major losses of Ca and base cations from the soil occur via leaching driven by atmospheric deposition of SO_4^{2-} and NO_3^- (e.g., Likens et al., 1996), leaching by naturally produced organic anions (Johnson et al., 1994b), and sequestration in forest vegetation, most importantly in the bole wood of long-lived trees (Johnson et al., 1994b; Likens et al., 1998). Inputs at low elevations in base-poor forest soils of the northeastern United States are primarily from mineral

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weathering, which can contribute >90% of Ca inputs (e.g., Likens and Bormann, 1995; Bailey et al., 1996). Specifically, Ca in Adirondack forest soils is released through the dissolution of Ca-bearing minerals in the till (April and Newton, 1992), which in the network of our plots are primarily calcic feldspars (J.E. Bedison and G.I. Omar, unpublished data, 2006). Mycorrhizal weathering of apatite can also be an important source of Ca to forest vegetation in northeastern U.S. forests (Blum et al., 2002; Yanai et al., 2005).

In high-elevation forests (≥ 900 m) throughout the Adirondacks, atmospheric inputs of base cations are important in regulating the base status of soils (Gbondo-Tugbawa and Driscoll, 2003) and contribute $\sim 50\%$ of the Ca currently in the forest floor (Miller et al., 1993). Several studies have also indicated that base cation deposition in precipitation has decreased across the northeastern United States in recent decades (e.g., Hedin et al., 1994; Likens et al., 1996; Gbondo-Tugbawa and Driscoll, 2003), which Likens et al. (1998) estimated was responsible for $\sim 20\%$ of the apparent depletion of soil Ca at the Hubbard Brook Experimental Forest (HBEF) in New Hampshire between 1940 and 1995. From remeasurements of Ca in organic horizons of high-elevation spruce–fir stands in the Adirondacks, Johnson et al. (2008a) showed a significant decrease in dilute-acid-extractable Ca during >70 yr, $\sim 25\%$ of which was probably attributable to SO_4^{2-} leaching.

Although widespread losses of Ca and base cations from forest soils in northeastern North America are suspected (e.g., Likens et al., 1996; Schaberg et al., 2001; Driscoll et al., 2003; Johnson et al., 2008a; Warby et al., 2009), forest soil Ca is not necessarily being depleted in all pools at all sites at high rates in the Adirondack region. Johnson et al. (1994a) reported that Ca concentrations and contents decreased in organic and E horizons but did not change between 1932 and 1984 in the B and C horizons of Adirondack forest soils. The same study also concluded that in a mixed hardwood–softwood forest near Newcomb, NY, total Ca loss from the soil could be accounted for by biomass uptake between 1930 and 1984. Moreover, using the NuCM model, Liu et al. (1992) predicted only minor future losses of available Ca during a 65-yr interval at the Huntington Forest in the central Adirondacks under several atmospheric deposition scenarios. Given the lack of recent empirical data, it is unclear what changes, if any, have occurred in the Ca status of mineral horizons in recent decades.

The objectives of this investigation were to quantify dilute-HCl-extractable Ca concentrations and content in organic and mineral horizons (to the bottom of the root zone) at three different times—1930 to 1932, 1984, and 2005/6—in a network of 54 permanent forest plots located throughout the Adirondack Mountains in the state of New York. Following previous work, we hypothesized that (i) extractable Ca in organic horizons decreased significantly between 1932 and 2005/6, (ii) Ca in the upper mineral soil decreased between all samplings, and (iii) Ca in deeper mineral horizons did not change with time.

MATERIALS AND METHODS

Site Description

The 54 permanently marked forest sites sampled in this investigation were located in the 2.5 million ha Adirondack Forest Preserve in New York (Fig. 1). This network of sites has been described in greater detail in Andersen (1988) and has been used in several comparative studies of vegetation and soil chemistry (Andersen, 1988; Johnson et al., 1994a, 2008a; Bedison et al., 2007; Bedison and Johnson, 2009; Bedison and McNeil, 2009). Represented in this network of plots are stands of northern hardwood (NH, $n = 20$), red and white pines with various associated hardwoods (PW, $n = 9$), high-elevation spruce–fir (SF, $n = 11$), and an intensively sampled 70-ha hardwood–softwood stand near Newcomb, NY (NC, $n = 14$). The main species in the NH plots were American beech (*Fagus grandifolia* Ehrh.), red maple (*Acer rubrum* L.), sugar maple (*Acer saccharum* Marsh.), and yellow birch (*Betula alleghaniensis* Britton), with red spruce (*Picea rubens* Sarg.) and eastern hemlock [*Tsuga canadensis* (L.) Carr] as the important associated conifers. The pine-dominated plots contained eastern white pine (*Pinus strobus* L.), red pine (*Pinus resinosa* Ait.), or both, with oaks (*Quercus* spp.) and various other hardwoods as associated species. The subalpine spruce–fir stands were located at elevations ≥ 900 m and were dominated by red spruce, paper birch [*Betula papyrifera* var. *cordifolia* (Regel) Fern.] and balsam fir [*Abies balsamea* (L.) Mill.]. The NC site had been used for thinning experiments until the 1950s (Andersen, 1988) and had a similar vegetation composition to the NH plots but

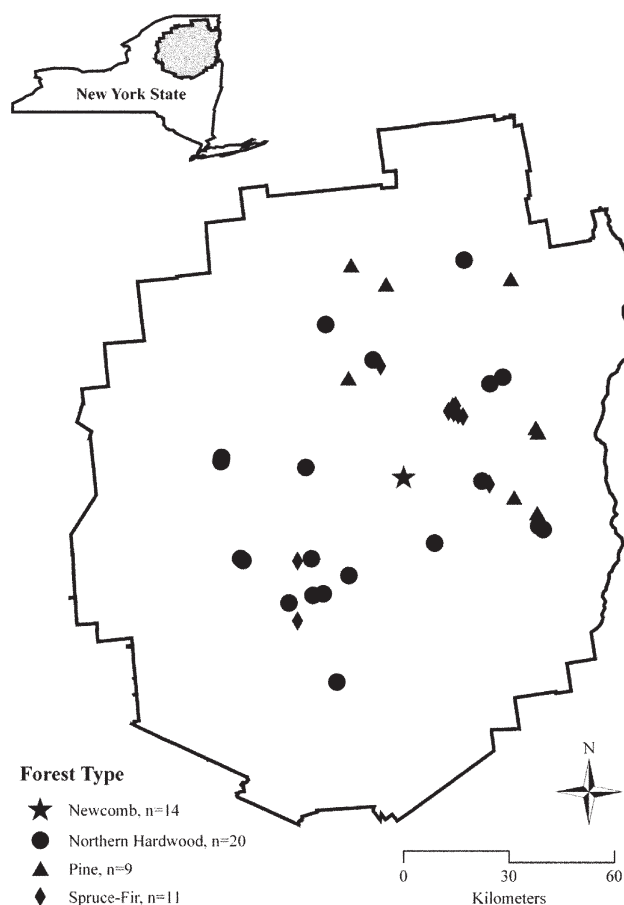


Fig. 1. Location of the study sites in the Adirondack Mountains.

with a substantial amount of eastern hemlock in some plots. Past episodes of fire, logging, blowdown, agricultural use, and severe red spruce mortality were easily detected disturbances in these stands (Bedison et al., 2010), as they typically are across the Adirondacks.

In the original investigation, Heimburger (1933) was interested in establishing “forest types” of the Adirondack region. He located his sampling sites in representative mature forest on well-drained soils. Consequently, the sites were neither randomly distributed nor systematically located throughout the Adirondack Park, although well-drained soils represent ~80% of the park area (Adirondack Park Agency, 2001). Therefore, we confined our conclusions to this network of plots. We are unaware, however, of any reasons why this set of forest plots might be different from a stratified random sample of Adirondack forests growing on well-drained sites.

Soil Sampling

Organic and mineral horizons were qualitatively sampled from an exposed pit face in 1930 to 1932 (Heimburger, 1933) and in 1984 (Andersen, 1988; Johnson et al., 1994a); however, no samples remain from the 1930 to 1932 investigation. Archived, air-dry samples from the 1984 investigation (Andersen, 1988) were stored in glass jars, although some of those samples had been exhausted before 2005.

In the summers of 2005/6, soils were sampled by excavating a 0.5- by 0.5-m quantitative pit at each site (*sensu* Hamburg, 1984). Pit locations were randomly located either within or directly adjacent to the permanently marked plot. Because the 1984 investigation noted the sampling locations relative to the plot centers (Andersen, 1988), care was taken to avoid sampling in the immediate vicinity of those locations.

Organic horizons (Oe and Oa) were weighed and subsampled in the field. We excavated and weighed rocks, roots, and mineral soil in 0- to 10-, 10- to 20-, and >20-cm depth intervals. The >20-cm interval was excavated to the bottom of the rooting zone, which was Andersen's (1988) functional definition of the bottom of the B horizon. Well-mixed subsamples of each mineral horizon (~2–3 kg) were collected. All soils were air dried in the laboratory. Following the procedures of Heimburger (1933) and Andersen (1988), air-dry soils were sieved according to horizon (Oe horizon, 5 mm; Oa horizon, 2 mm; mineral soil, 1 mm) and stored in jars before analysis.

As the soil sampling method was different in 2005/6, we pooled and partitioned measurements from the genetic mineral horizons in the 1932 and 1984 studies into the appropriate depth intervals as represented in the 2005/6 sampling. The depth to the bottom of the B horizon at each site was generally different between years, so for comparisons of Ca content, we truncated the >20-cm increment at the shallowest depth measured in the three studies for each site. In 2005/6, we determined the bulk density (D_b) of the organic and mineral layers and showed that D_b was highly correlated with the organic matter concentration measured as mass loss-on-ignition (LOI; Bedison et al., 2010). We used a polynomial regression equation fitted to those data to estimate the D_b from the LOI measurements reported in the 1932 and 1984 studies (Bedison et al., 2010). More detailed descriptions of soil sampling procedures used in this investigation can be found in Andersen (1988) and Bedison and Johnson (2009).

Calcium Measurements

Following the methods of Heimburger (1933), sieved samples from the 1984 and 2005/6 studies were dried by heating ~2.0 g of each sample at 95°C for 12 h. The oven-dry soil was combined with 20 mL of 0.2 mol L⁻¹ HCl, shaken for 4 h, and then filtered. The filtrate was evaporated to dryness and heated at 550°C for 12 h to destroy any remaining organic matter. The combusted filtrate was then dissolved in 20 mL of 0.2 mol L⁻¹ HCl and analyzed for Ca using an inductively coupled plasma–atomic emission spectrometer (ICP-AES; Spectro Analytical Instruments Inc., Mahwah, NJ) using standard procedures. Concentrations of Ca in 1932 were determined with a calcium oxalate titration procedure (Heimburger, 1933). No other extractable base cations (e.g., Mg, K) were measured in that study, thus long-term comparisons are restricted to Ca. Using the horizon thickness (T_h) and the measured or estimated D_b , Ca concentrations were converted to Ca content for each soil horizon in each year. Following Heimburger (1933), all results are reported on an oven-dry mass basis.

Comparability of Methods

Comparability of the Ca determination methods is one of the most critical aspects of this study. With regard to possible errors in the Ca determination, Andersen (1988) provided data related to the potential for artifacts related to how the laborious wet-chemical method used by Heimburger (1933) for Ca determination might be subject to errors from the lack of Fe and Al removal and the loss of Ca during the many steps of washing precipitates from filter paper to beakers. The results in Andersen (1988) and Johnson et al. (2008a) showed that if the 1930s calcium oxalate method is done carefully, the results for extractable Ca in both mineral and organic horizons are the same as those obtained using an atomic absorption spectrophotometer (AAS). As the AAS used to generate those comparisons is no longer available, forest floor Ca concentrations measured in an unrelated investigation in 1979 with the AAS were compared with the results obtained with the ICP-AES, using the original samples from that study to evaluate comparability (Johnson et al., 2008a). The results from that comparison revealed that Ca determined with the ICP-AES matched the results obtained with the AAS approximately 27 yr prior. We believe that the Ca values reported for the 1984 and 2005/6 samples here are comparable to those obtained by Heimburger (1933).

The biological relevance of Ca extracted by 0.2 mol L⁻¹ HCl is not known, but the amounts of Ca extracted are 1.6 times greater in organic horizons than Ca extracted by 1 mol L⁻¹ NH₄Cl (Johnson et al., 2008a) and similar in magnitude to the total Ca values obtained by acid digestion of the forest floor (A.H. Johnson, unpublished data, 2009). In mineral horizons, the HCl-extractable Ca values measured by Andersen (1988) varied by horizon and were <10% of the total Ca values measured by complete digestion of the soil (LiBO₃ fusion and digestion). Accordingly, we believe that the dilute acid procedure used here extracts more Ca than would be released in a neutral salt extraction (e.g., 1 mol L⁻¹ NH₄Cl), but in mineral horizons it does not appear to be dissolving large quantities of the Ca-bearing minerals.

Statistical Analyses

We calculated the Ca contents (kg ha^{-1}) and concentrations ($\text{cmol}_c \text{ kg}^{-1}$ oven-dry soil) of organic and mineral horizons in 1932, 1984, and 2005/6. We also calculated the Ca concentration on an organic matter (OM) basis ($\text{cmol}_c \text{ kg}^{-1}$ OM) to allow for potential sampling biases, especially in the organic horizons. We used paired-comparison tests to evaluate the significance of differences in extractable Ca between sampling dates and a Shapiro–Wilk test to determine if the data were normally distributed. Data that were normally distributed, either untransformed or \log_{10} transformed, were compared using a paired *t*-test. A Wilcoxon's signed-ranks test was used when the transformed data were not normally distributed. Because the majority of data were not normally distributed, we report both mean and median values. Rates of change between years were determined using median values. Univariate linear regression analysis on untransformed data was used to evaluate the relationship between OM concentration and Ca content in each sampling. All data were analyzed with JMP (version 7.0.1, SAS Institute, Cary, NC) and statistical significance was evaluated at $P \leq 0.05$ ($\alpha = 0.05$).

RESULTS AND DISCUSSION

Extractable Calcium for Pooled Data: 1932, 1984, and 2005/6

Extractable organic horizon (Oe + Oa) Ca contents (Table 1) and concentrations (Fig. 2) decreased significantly among the three sampling dates, and the median Ca content decreased at

a rate of $8.1 \text{ kg Ca ha}^{-1} \text{ yr}^{-1}$ between 1932 and 2005/6. This long-term rate of change in Ca content ($-1.1\% \text{ yr}^{-1}$) was consistent with the rate of change, $0.9\% \text{ yr}^{-1}$, that was necessary to detect a significant difference in Ca content of the forest floor at the HBEF (Yanai et al., 1999). Moreover, between 1984 and 2005/6, the organic horizon Ca concentration decreased significantly, and the median Ca content was significantly lower in 2005/6 than in 1984 (-32 and $-1.5\% \text{ yr}^{-1}$, respectively). This trend was comparable to the decrease in exchangeable Ca in the organic horizons (-36%) reported for Adirondack watersheds surveyed under the Direct/Delayed Response Program between 1984 and 2001 (Warby et al., 2009).

Similar to results reported by Likens et al. (1998), univariate linear regression analysis indicated that the extractable Ca content of these Adirondack forest soils was significantly correlated with the OM concentration. Between 1932 ($R^2 = 0.38$, $m = 0.45$, $P < 0.0001$) and 1984 ($R^2 = 0.36$, $m = 0.10$, $P < 0.0001$) there was a large decrease in the amount of Ca associated with the organic matter, as indicated by a decrease in regression slope (m) between the two sampling dates. Moreover, there was a slight decrease in Ca associated with the soil OM between 1984 and 2005/6 ($R^2 = 0.47$, $m = 0.09$, $P < 0.0001$). Although this decrease in m was not significant, it provides further evidence of continued soil Ca depletion. It is clear that the soil Ca content is strongly associated with the OM concentration of Adirondack forest soils, thus a change in the OM concentration may be spuriously interpreted as a change in the soil Ca content.

To account for this possibility, Ca concentrations were also reported on an OM basis. We found that the Ca concentration ($\text{cmol}_c \text{ kg}^{-1}$ OM) in organic horizons also decreased significantly among the three sampling periods (Fig. 2). Collectively, these results suggest that Ca depletion in organic horizons has been sustained throughout the 20th century in the Adirondack region and was not a result of sampling differences between investigations. While most of the Ca loss could be due to leaching and vegetation uptake and sequestration, it is possible that the Ca content of the litter has also decreased, though there are no data that would help identify temporal changes in litterfall Ca concentration or inputs.

In the upper mineral horizons (i.e., 0–10 and 10–20 cm), the Ca content (Table 1) and concentration (Table 2) decreased during the intervals between the sampling periods and there was significantly less Ca in 2005/6 than in 1932. The Ca concentration ($\text{cmol}_c \text{ kg}^{-1}$ OM) also decreased significantly in the 0- to 10-cm mineral horizons between 1932 and 1984. This is consistent with the trends in E horizon Ca depletion reported by Johnson et al. (1994a), as the E horizons were within the 0- to 10-cm mineral soil. Furthermore, this trend of decreasing Ca continued between 1984 and 2005/6 (Table 2). In contrast to the upper mineral horizons, the

Table 1. Organic horizon, mineral horizon and whole-profile Ca content in 1932, 1984, and 2005/6 for the pooled data. Years for a given horizon followed by different letters are significantly different ($P \leq 0.05$).

Horizon	<i>n</i>	Year	Mean	Percentile		
				25	Median	75
kg Ca ha^{-1}						
Oe horizon	47	1932 a	303.1	128.1	261.3	388.4
		1984 b	101.9	69.0	97.5	129.5
		2005–2006 c	55.1	23.4	47.3	69.1
Oa horizon	50	1932 a	726.1	197.0	417.5	1042.9
		1984 b	344.6	67.7	150.9	391.0
		2005–2006 c	165.1	41.4	110.7	238.1
Total organic†	51	1932 a	993.5	427.6	763.6	1402.6
		1984 b	431.8	136.2	238.7	473.1
		2005–2006 c	209.6	84.5	163.3	292.3
0–10 cm	54	1932 a	409.8	110.3	324.6	574.3
		1984 b	267.9	38.0	79.2	326.9
		2005–2006 b	175.6	46.9	107.7	233.8
10–20 cm	54	1932 a	310.1	34.8	166.5	486.0
		1984 a	289.5	49.8	116.0	314.5
		2005–2006 b	144.9	30.4	70.3	212.5
>20 cm	54	1932 a	301.7	0.0	62.4	372.7
		1984 a	485.7	21.6	175.7	657.0
		2005–2006 a	373.4	15.4	151.2	447.5
Total mineral soil	54	1932 ab	1019.5	163.0	685.0	1456.4
		1984 a	1074.2	155.9	619.8	1370.2
		2005–2006 b	705.6	138.7	484.2	1142.6
Whole profile‡	54	1932 a	1948.2	1200.2	1575.8	2490.7
		1984 b	1450.9	497.1	865.8	1499.9
		2005–2006 c	891.9	265.7	571.1	1226.8

†Total organic = Oe + Oa horizons.

‡Whole profile = total organic + total mineral.

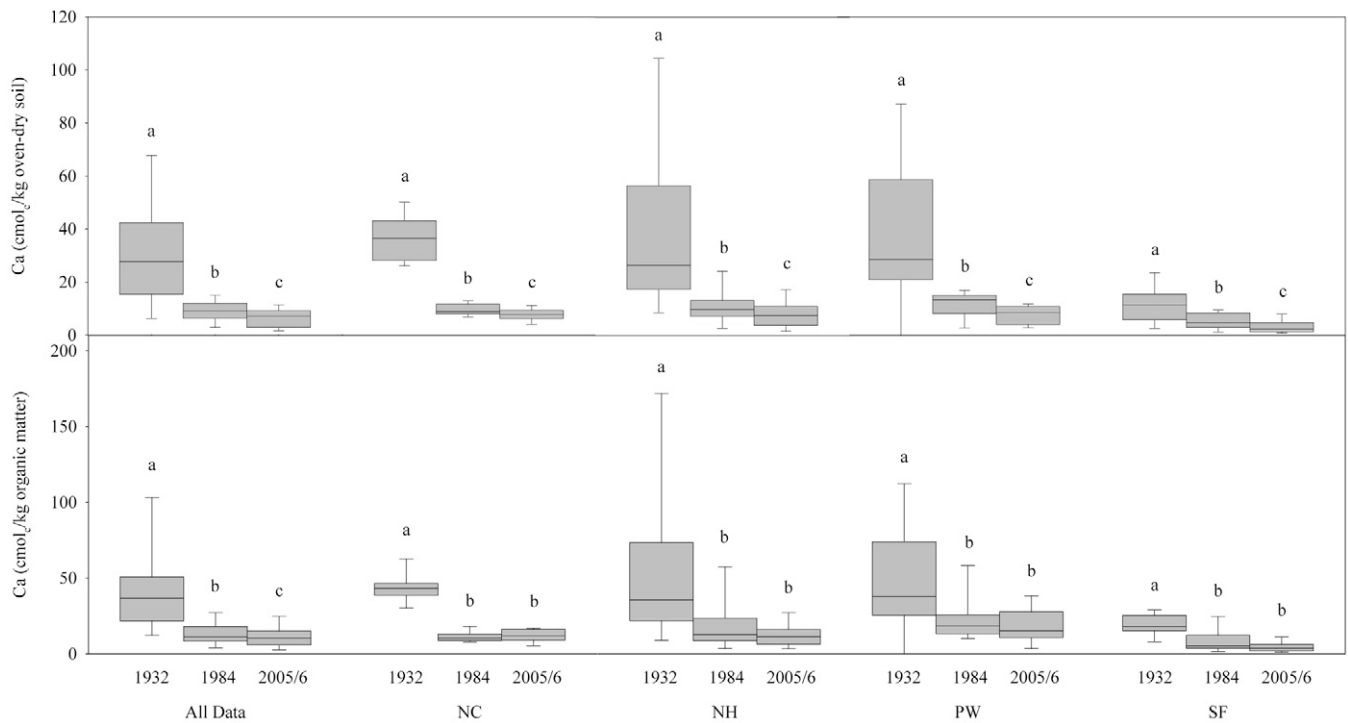


Fig. 2. Organic horizon (Oe + Oa) Ca concentrations for all data, Newcomb (NC), northern hardwood (NH), pine-dominated (PW), and spruce-fir plots (SF) in 1932, 1984, and 2005/6. The boxes represent the upper and lower quartiles while the bar in the center represents the median. The whiskers represent the 10th and 90th percentiles of the data. For a given forest type, years not sharing the same letter are significantly different ($P \leq 0.05$).

Ca concentration and content in the >20-cm mineral soil did not change during any interval (Tables 1 and 2).

Several other decadal-scale investigations of forest soils in the northeastern United States have reported depletion of Ca in combined organic and mineral horizon pools. Bailey et al. (2005) reported moderate ($12.0 \text{ kg ha}^{-1} \text{ yr}^{-1}$) to excessive depletion rates of exchangeable Ca ($24.4\text{--}197.2 \text{ kg ha}^{-1} \text{ yr}^{-1}$) in genetic horizons of rocky forest soils of the Allegheny Plateau between 1967 and 1997. The mechanism(s) that could cause such rapid rates of Ca loss is unknown, as the rates at the upper end of the range exceed reasonable rates of leaching and sequestration in biomass (Johnson, 2005). Exchangeable Ca also decreased in mixed hardwood-softwood forest soils in central Ontario at rates of 1.9 to $16.9 \text{ kg ha}^{-1} \text{ yr}^{-1}$ between 1983 and 1999 (Watmough and Dillon, 2003). Excluding one plot with an unexplained increase, exchangeable Ca also decreased significantly between 1982 and 2004 in the Walker Branch Watershed in Tennessee at a rate of $12.5 \text{ kg ha}^{-1} \text{ yr}^{-1}$ (Johnson et al., 2008b). At the HBEF, soil Ca decreased between 1982 and 1992 at a rate of $10.4 \text{ kg ha}^{-1} \text{ yr}^{-1}$ (Likens et al., 1998). In the 54 Adirondack forest plots sampled in this investigation, the median whole-profile extractable Ca content was significantly less between all three sampling dates and decreased at a rate of $\sim 13.6 \text{ kg Ca ha}^{-1} \text{ yr}^{-1}$ across all intervals. Given that the Ca concentration in both the organic (Fig. 2) and upper mineral horizons (Table 2) continued to decrease between 1984 and 2005/6, our results indicate that, despite decreases in airborne SO_4^{2-} deposition in recent decades and the concomitant reduction in the Ca leaching potential, losses of Ca from

Table 2. Concentration of soil Ca on both a whole-soil and an organic matter (OM) basis in the pooled data in 1932, 1984, and 2005/6. For a horizon, years not sharing the same letter are significantly different ($P \leq 0.05$).

Depth	Basis	Year	Mean	Percentile			
				25	Median	75	
cm							
cmol _c kg ⁻¹							
0–10	OM	1932 a	56.8	4.8	31.7	71.2	
		1984 b	18.2	3.5	9.2	23.6	
		2005–2006 c	10.7	3.6	7.3	14.9	
	whole soil	1932 a	2.7	1.1	2.0	3.7	
		1984 b	1.9	0.3	0.7	2.2	
		2005–2006 b	1.5	0.3	0.9	1.8	
	10–20	OM	1932 a	39.1	2.5	12.2	36.2
			1984 a	20.6	3.5	8.3	25.1
			2005–2006 b	9.5	2.4	6.7	14.2
whole soil		1932 a	2.5	0.7	1.1	3.6	
		1984 a	2.2	0.4	1.0	2.3	
		2005–2006 b	1.3	0.2	0.7	1.9	
0–20	OM	1932 a	47.9	3.9	26.7	56.2	
		1984 b	19.4	3.7	9.3	24.0	
		2005–2006 c	10.1	3.5	8.1	13.4	
	whole soil	1932 a	2.6	0.8	1.5	3.6	
		1984 a	2.1	0.4	0.9	2.5	
		2005–2006 b	1.4	0.3	0.9	1.8	
>20	OM	1932 a	27.1	0.0	7.1	36.2	
		1984 a	32.8	3.4	14.6	45.2	
		2005–2006 a	21.8	1.4	11.0	34.7	
	whole soil	1932 a	1.6	0.0	1.1	2.6	
		1984 a	2.3	0.3	1.4	2.7	
		2005–2006 a	1.6	0.2	0.9	2.5	

the root zone have exceeded combined inputs from atmospheric deposition and mineral weathering in these Adirondack forests since 1984.

Changes in Soil Calcium in the Northern Hardwood and Newcomb Plots

Northern hardwood forests and the mixed conifer-hardwood forests represented in the NC plots dominate the Adirondack landscape (e.g., McMartin, 1994). The Ca contents and concentrations were significantly less in organic horizons in both NH and NC plots in 2005/6 compared with 1932 (Table 3 and Fig. 2, respectively). There were significant differences in

the Ca concentration of the 0- to 20-cm mineral soil in the NH plots but not in the NC plots (Fig. 3). The >20-cm mineral horizon Ca content and concentration were not different between any sampling times in either NC or NH plots. During the 74-yr interval, the median whole-profile Ca content decreased at similar rates in both the NC (13.9 kg ha⁻¹ yr⁻¹) and NH (12.8 kg ha⁻¹ yr⁻¹) plots. These long-term rates of Ca depletion were similar to soil solution fluxes of Ca, 4.5 to 14.8 kg ha⁻¹ yr⁻¹, measured between 1983 and 1992 in northern hardwood forests at the HBEF in New Hampshire (Johnson et al., 2000) and to mass balance estimates of Ca loss from the exchangeable pool in a southern Piedmont forest (12.7 kg ha⁻¹ yr⁻¹; Huntington

et al., 2000). Moreover, whole-profile Ca content was less in 2005/6 than in 1984, although not significantly. Our results indicate that losses from the organic and upper mineral horizons in these forests have been greater than combined inputs since the early 1930s and that the losses of Ca deeper in the profile were balanced by mineral weathering inputs and possibly by leaching of Ca from the upper soil horizons.

Pine-Dominated Plots

Little research has focused on nutrient cycling and the soil Ca status in pine-dominated forests in the Adirondack Mountains, perhaps owing to the limited spatial extent of this forest type (e.g., McMartin, 1994). The extractable Ca content (Table 3) and concentration (Fig. 2 and 3) were significantly greater in all soil pools in 1932 than 1984 or 2005/6. The loss rate of median whole-profile Ca content in the PW plots between 1932 and 1984 (27.5 kg ha⁻¹ yr⁻¹) was greater than that between 1984 and 2005/6 (18.1 kg ha⁻¹ yr⁻¹), thus indicating that Ca loss in the PW plots has been relatively high but may have slowed in recent decades.

Although the Ca requirements of *Pinus* spp. and other conifers are generally less than those of deciduous species in eastern U.S. forests (e.g., Johnson and Lindberg, 1992), Ca sequestration in aggrading biomass may account for much of the measured soil Ca depletion between 1984 and 2005/6 in the PW plots. Knoepp and Swank (1994) measured a Ca depletion rate of 12.3 kg ha⁻¹ yr⁻¹ in the A horizons of a white pine watershed at the Coweeta Hydrological Laboratory in North Carolina between 1970 and 1990 and reported that biomass accumulation

Table 3. Total organic, total mineral, and whole-profile Ca contents for Newcomb (NC), northern hardwood (NH), pine-dominated (PW), and spruce-fir (SF) sites in 1932, 1984, and 2005/6. For a given horizon, years within the same forest type followed by different letters are significantly different ($P \leq 0.05$).

Forest type	Horizon	Year	Mean	Percentile		
				25	Median	75
kg Ca ha ⁻¹						
NC	total organic†	1932 a	1371.4	716.9	1358.5	2044.1
		1984 b	405.3	215.2	275.1	683.6
		2005–2006 b	245.2	158.3	248.6	303.1
	total mineral	1932 a	554.5	238.9	565.3	822.5
		1984 a	883.4	487.3	709.4	1174.2
		2005–2006 a	763.8	462.8	627.0	1142.6
	whole profile‡	1932 a	1888.9	1290.5	1685.3	2416.8
		1984 b	1169.1	821.6	1002.6	1456.2
		2005–2006 b	964.2	534.6	658.9	1343.8
NH	total organic	1932 a	930.4	386.9	607.2	979.5
		1984 b	498.1	206.7	227.5	489.0
		2005–2006 b	273.6	88.7	215.2	385.3
	total mineral	1932 a	1430.2	361.2	962.9	1953.7
		1984 a	1549.7	271.2	703.6	1844.5
		2005–2006 a	922.0	344.8	592.2	1479.8
	whole profile	1932 a	2221.0	705.3	1750.4	3104.4
		1984 ab	1973.2	514.2	815.3	2114.8
		2005–2006 b	1154.6	427.7	804.9	2069.6
PW	total organic	1932 a	428.7	216.8	261.3	614.6
		1984 a	217.6	99.1	128.7	267.7
		2005–2006 b	72.4	28.0	66.5	112.2
	total mineral	1932 a	1636.0	960.0	1332.9	2315.7
		1984 ab	1049.9	159.5	747.7	1693.9
		2005–2006 b	938.9	157.6	453.8	1974.3
	whole profile	1932 a	2064.7	1272.7	2304.6	2499.4
		1984 ab	1267.5	258.6	876.4	1961.7
		2005–2006 b	1011.3	227.3	478.3	2061.4
SF	total organic	1932 a	1071.9	801.1	1018.4	1407.5
		1984 b	538.0	80.6	269.2	554.8
		2005–2006 b	177.5	83.4	146.9	318.3
	total mineral	1932 ab	360.4	0.0	103.9	195.9
		1984 a	472.3	104.4	153.4	377.0
		2005–2006 b	47.2	0.0	0.0	135.3
	whole profile	1932 a	1432.3	809.5	1308.2	1521.3
		1984 a	1010.3	170.4	683.9	866.2
		2005–2006 b	224.7	107.8	163.3	318.3

† Total organic = Oe + Oa horizons.

‡ Whole profile = total organic + total mineral.

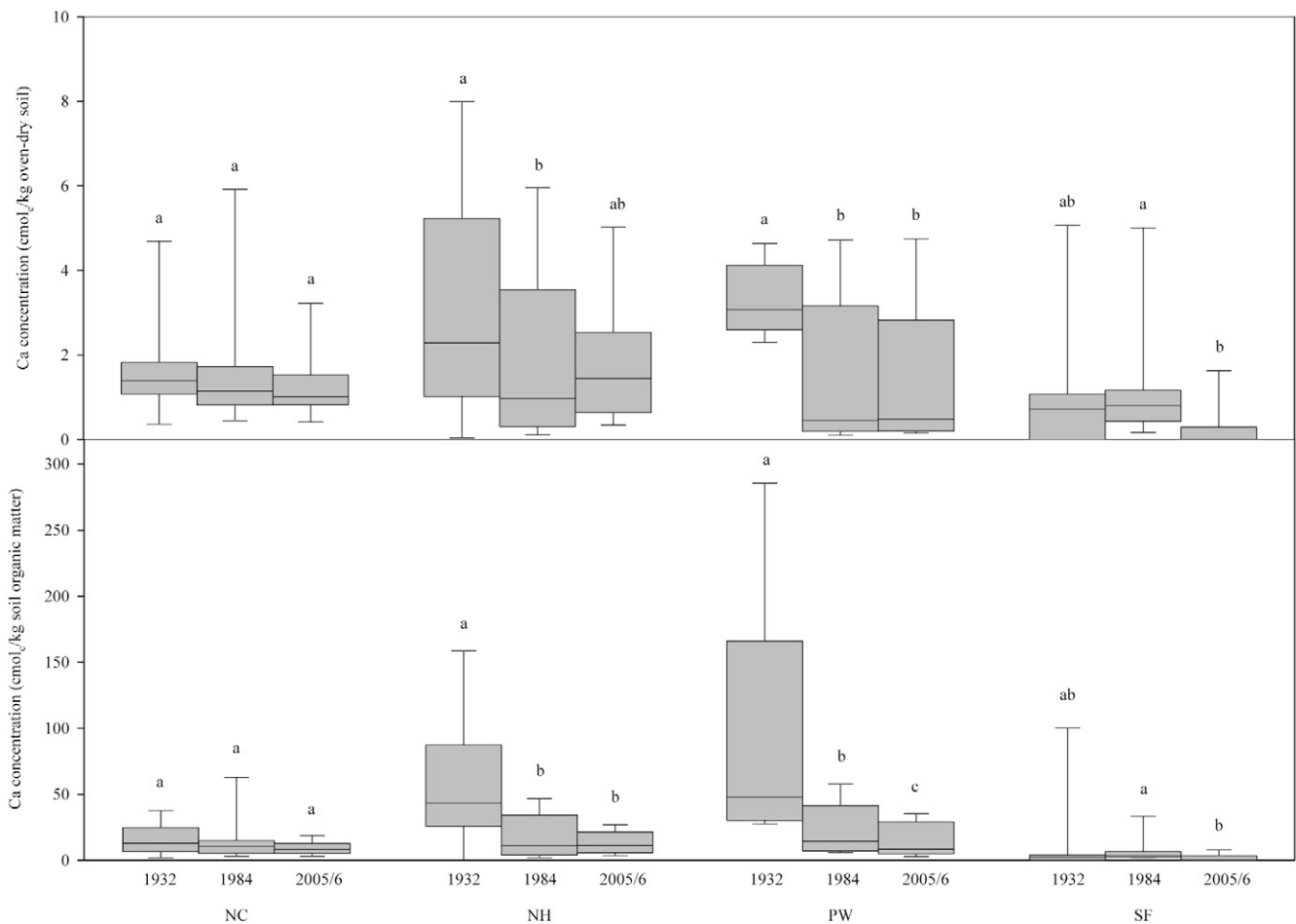


Fig. 3. Mineral horizon (0–20 cm) Ca concentration in Newcomb (NC), northern hardwood (NH), pine-dominated (PW), and spruce–fir (SF) plots in 1932, 1984, and 2005/6. The boxes represent the upper and lower quartiles while the bar in the center represents the median. The whiskers represent the 10th and 90th percentiles of the data. For a given forest type, years not sharing the same letter are significantly different ($P \leq 0.05$).

rather than leaching was the primary mechanism. Between 1984 and 2004, data in Bedison et al. (2007) indicated that the live biomass in the PW plots was aggrading much more rapidly (change in basal area = $0.5 \text{ m}^2 \text{ ha}^{-1} \text{ yr}^{-1}$) than in the NH and SF plots. The whole-profile Ca depletion rate measured between 1984 and 2005/6 was $18.1 \text{ kg ha}^{-1} \text{ yr}^{-1}$. In agreement with the results of Knoepp and Swank (1994), soil Ca depletion rates measured for the PW plots in this investigation suggest that Ca sequestration in biomass may be an equally important factor as leaching in soil Ca depletion in Adirondack forests where biomass is aggrading rapidly.

Changes in Soil Calcium in High-Elevation Spruce–Fir Plots

Due to the high spatial variability in high-elevation SF forest soils, comparisons of soil chemistry between years should be interpreted with caution. Because seven of the SF sites sampled in 1932 had mineral soil, all 11 sites had mineral soil in 1984, but only five sites in 2005/6 had mineral soil, interpreting temporal comparisons in mineral horizon Ca content in the SF plots is potentially misleading. Despite potential sampling biases (e.g., Yanai et al., 2000), the Ca concentration ($\text{cmol}_c \text{ kg}^{-1} \text{ OM}$) in the organic horizons of the SF sites decreased during all intervals and was significantly less in 2005/6 than in 1932 (Fig. 2). The long-

term (74-yr) Ca loss rate determined for the organic horizons in the SF plots ($11.8 \text{ kg ha}^{-1} \text{ yr}^{-1}$) was greater than the annual Ca loss rates determined in a 4-yr biogeochemical study of a SF forest on Whiteface Mountain in New York ($8.4 \text{ kg ha}^{-1} \text{ yr}^{-1}$; Johnson et al., 1994b), and that reported for the forest floor at these sites by Johnson et al. (2008a). Furthermore, the median organic horizon Ca depletion between 1984 and 2005/6 ($5.6 \text{ kg ha}^{-1} \text{ yr}^{-1}$) was lower than both the 52-yr rate and the depletion rate measured between 1932 and 1984 ($14.4 \text{ kg ha}^{-1} \text{ yr}^{-1}$). Despite apparent reductions in the rate of Ca loss, relative losses between 1984 and 2005/6 ($2.1\% \text{ yr}^{-1}$) were greater than those between 1932 and 1984 ($1.4\% \text{ yr}^{-1}$). These results indicate that Ca depletion has continued in those horizons with time and the soil Ca pool has continued to be reduced.

Controls on Calcium Loss

The principal processes that regulate the soil Ca content in Adirondacks forests are (i) atmospheric inputs, (ii) sequestration of Ca in living biomass, (iii) leaching due to organic anions, SO_4^{2-} and in some cases NO_3^- , and (iv) perhaps Ca return in litter. We expected those processes to have different rates in different forests because some forests are accumulating biomass faster than others (e.g., Bedison et al., 2007), the delivery of SO_4^{2-}

and NO_3^- varies from west to east (Ollinger et al., 1993; Ito et al., 2002; McNeil et al., 2007) and from low to high elevation (Miller et al., 1993), the production of organic acids varies with temperature and litter quality, and the base cation content and input of litter vary by species (e.g., Johnson and Lindberg, 1992). The best way to accurately estimate the various influences on Ca depletion is by obtaining plot-level fluxes, which was beyond the scope and capacity of this investigation. There are, however, data available from some Adirondack biogeochemical budget studies that are relevant to the observed depletion rates.

The measured rates of soil Ca loss from sequestration in biomass ($2.7\text{--}8.2 \text{ kg ha}^{-1} \text{ yr}^{-1}$; Andersen, 1988; Johnson and Lindberg, 1992; Johnson et al., 1994a) and net leaching from the rooting zone ($5.0\text{--}12.2 \text{ kg ha}^{-1} \text{ yr}^{-1}$; Johnson and Lindberg, 1992; Friedland and Miller, 1999) from detailed nutrient-cycling investigations throughout Adirondack forests constrain reasonable estimates of Ca loss from the soil pool to between ~ 8 and $20 \text{ kg ha}^{-1} \text{ yr}^{-1}$. Moreover, considering the substantial decrease in atmospheric Ca deposition since the 1960s (e.g., Barnes et al., 1982; Oehlert, 1984; Likens et al., 1998) and that weathering rates in the Adirondacks are typically low ($1.8\text{--}3.0 \text{ kg ha}^{-1} \text{ yr}^{-1}$; April et al., 1986; Andersen, 1988), net outputs of Ca from the soil pool are probably greater than inputs in Adirondack forests. Given the rates of Ca input and loss reported for various Adirondack forest soils, the overall ($n = 54$) whole-profile Ca depletion rate measured here ($\sim 13.6 \text{ kg ha}^{-1} \text{ yr}^{-1}$) for all intervals seems reasonable. Without site-specific data, however, local variability in geology, vegetation, and deposition make it difficult to construct accurate Ca budgets. Future work in this network of plots is needed to measure nutrient budgets and assess the relative influence of various processes on the nutrient status of these forests soils.

CONCLUSIONS

Repeated measurements of dilute HCl-extractable Ca in 54 Adirondack forest soils in 1932, 1984, and 2005/6 revealed that Ca decreased significantly in organic and upper mineral horizons during both the long term (1932–2005/6) and more recent (1984–2005/6) intervals. The extractable Ca concentration and content of mineral horizons deeper than 20 cm did not change significantly and there was not a trend toward decreased Ca content in those horizons. This suggested that mineral weathering and inputs from overlying horizons mitigated Ca losses deeper in the profile but not in the upper soil horizons. Trends in Ca depletion in the NH, NC, and PW plots were similar to trends in the pooled data. In the high-elevation SF plots, variability in mineral horizons among samplings made estimates of whole-soil Ca depletion rates uncertain, but decreases in organic horizon Ca pools have been clearly established by this and related investigations. While we do not currently have site-specific data to construct detailed Ca budgets, the existing data from nutrient cycling investigations were consistent with the measured changes in Ca in this network of Adirondack forest soils. Our investigation indicated that the soil Ca status of Adirondack forests has

not stabilized in recent decades and that depletion has continued despite reductions in atmospheric S deposition. Moreover, sustained losses of soil Ca combined with depleted soil pools suggest that soil Ca monitoring efforts in these and similar northeastern U.S. forest soils should be continued and that the anticipated impacts of reduced atmospheric S inputs on reducing forest soil Ca loss across the Adirondacks are not yet apparent.

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