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Soil Sci. Soc. Am. J. 74:2187-2195

Published online 23 Aug. 2010

doi:10.2136/sssaj2009.0367

Received 28 Sept. 2009.

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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(April and Newton, 1992), which in the network of our plots are primarily calcic feldspars (J.E. Bedison and G.I. Omar, unpublished data, 2006).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 ~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 ~20% of the apparent depletion of soil Ca

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<sup>-1</sup> 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<sup>-1</sup> HCl and analyzed for Ca using an inductively coupled plasma–atomic emission spectrometer (ICP-AES; Spectro

## Statistical Analyses

We calculated the Ca contents ( $\text{kg ha}^{-1}$ ) 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  $P = 0.05$  ( $\alpha = 0.05$ ).







and  $\text{NO}_3^-$  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  $\sim 8$  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.1 (ile)-13 1(g)-0.6l8of C-10ntial de-he -1c8(r)8(e)0(2TJ 89(r)7.9(ie)-13.1(7,p)-0.48-640



