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Long-Term Effects of Acid Rain: Response and Recovery of a Forest Ecosystem

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Long-term data from the Hubbard Brook Experimental Forest, New Hampshire, suggest that although changes in stream pH have been relatively small, large quantities of calcium and magnesium have been lost from the soil complex and exported by drainage water because of inputs of acid rain and declines in atmospheric deposition of base cations. As a result, the recovery of soil and streamwater chemistry in response to any decreases in acid deposition will be delayed significantly.

Research on the effects of acid rain (1) in the United States and Europe has focused primarily on the biogeochemistry of sulfur, and to a lesser extent on that of nitrogen (2). This emphasis was because sulfuric acid is the dominant acid in precipitation throughout the eastern United States and Europe where acid rain is a serious environmental problem (3). Records since 1963 show that rain and snow in northeastern United States have had an average annual pH of 4.05 to 4.3 and that sulfuric acid contributed 55 to 75% of this measurable acidity (4). However, this focus on sulfur obscured the role of other elements in the regulation of the long-term, acid-base status of precipitation, soil, and surface waters, as well as linkages between element cycles.

After passage of the Clean Air Act in 1970, emissions of SO₂ and particulates were reduced, and concentrations of sulfate in surface waters in eastern North America declined (Fig. 1, A and B) (5-7). Despite such changes, surface waters throughout the region that are low in acid neutralizing capacity (ANC) (8) have not shown a commensurate recovery in pH and ANC. Additional reductions in emissions and concomitant decreases in the deposition of strong acids are expected as a result of the 1990 amendments to the Clean Air Act. However, critical questions remain: will forest and aquatic ecosystems recover from the effects of acid rain, and if so, at what rate?

The chemistry of bulk precipitation and stream water has been measured continuously at Hubbard Brook Experimental Forest (HBEF) since 1963. These data show that annual, volume-weighted concentrations of SO₄²⁻, Ca²⁺, Mg²⁺, and Na⁺ declined in both precipitation and stream water since 1963; K⁺ declined in precipitation. These declines in atmospheric deposition at HBEF followed declines in atmospheric emissions since 1970 (Fig. 1) (4, 5, 7, 9,

10). On the basis of mass balance calculations, between 77% and 85% of the decline in stream base cations C_B (11) can be explained by decreases in the atmospheric deposition of C_B (7). The magnitude of decline of C_B in stream water was similar to that in bulk precipitation (30 to 40 μeq liter⁻¹) after 1963 (Fig. 1D).

Before 1970 (12), concentrations of C_B generally were increasing in stream water in

response to high or increasing concentrations of strong acid anions (SO₄²⁻ + NO₃⁻) (13). Concentrations peaked in 1969 to 1970 (Fig. 1, B and D), coincident with elevated concentrations of NO₃⁻ in stream water (4), and then declined (Fig. 1B). Significantly, the time path of the relation between C_B and strong acid anions in stream water shows a hysteresis pattern (Fig. 2).

It is thought that acid rain began around 1950 to 1955 in the northeastern United States (14). Although they are the longest record in North America, the data from HBEF do not include this period. There are data, however, for Ca²⁺, Na⁺, and K⁺ in precipitation for 1955-1956 (15). Using the relation of Mg²⁺ to Ca²⁺ during 1963 to 1970, we estimated the Mg²⁺ concentration for 1955-1956 and thus a C_B concentration of 53.5 μeq liter⁻¹ (Fig. 1D), which is about five times the average value for 1984 to 1993 (Fig. 1D) (16). We also estimated the concentration of C_B, strong acid anions, and ANC in stream water in 1940 (Fig. 2) (17).

Calcium is the major cation in stream water at HBEF (4) and is indicative of C_B. We apportioned the ecosystem sources and sinks of Ca²⁺ (Fig. 3) from 1940 to 1963 using estimated values, and from 1963 to

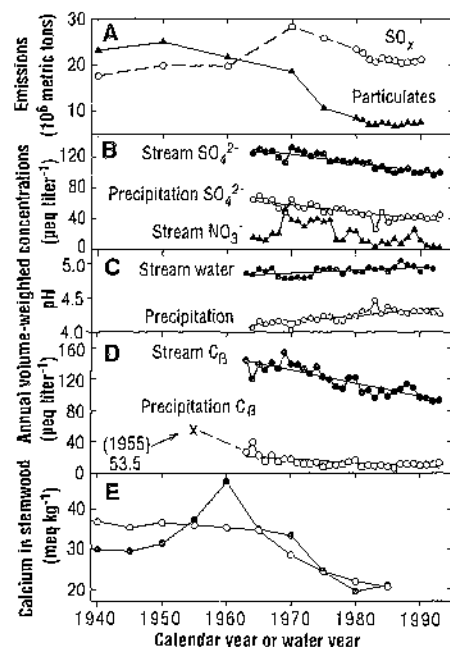


Fig. 1. Long-term trends in (A) emissions of SO₂ and particulates for the United States (28); (B) SO₄²⁻ and NO₃⁻ in stream water and SO₄²⁻ in bulk precipitation for Watershed 6 (W6) of the Hubbard Brook Experimental Forest; (C) pH of stream water and bulk precipitation for W6; (D) sum of base cation (C_B) concentrations in bulk precipitation and stream water for W6 [data for 1955-1956 (x) from (15)]; and (E) calcium concentrations in radial increments of stemwood of red spruce (*Picea rubens* Sarg.) from Carnels Hump Mountain, Vermont (●), and Crawford Notch, New Hampshire (○) (25). Solid trend lines in (B), (C), and (D) indicate a significant linear regression ($P < 0.05$ for a larger F value). Years span calendar years for (A) and (E) and water years for (B) through (D).

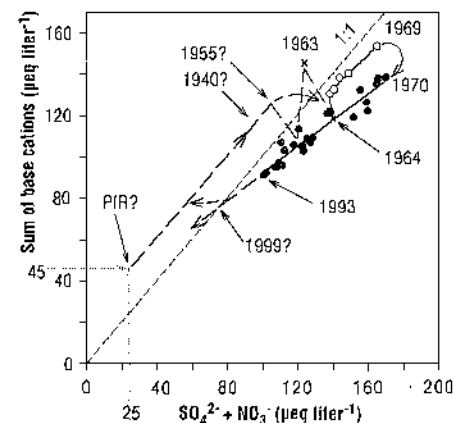


Fig. 2. Relation between annual, volume-weighted average concentration of the sum of base cations (C_B) and the sum of SO₄²⁻ + NO₃⁻ for stream water from Watershed 6 of the Hubbard Brook Experimental Forest from 1964-1965 through 1993-1994. The linear regression for data from 1965-1966 through 1969-1970 (○) is $Y = 0.83a + 17.4$ ($r^2 = 0.98$), and for 1970-1971 through 1993-1994 (●) is $Y = 0.63a + 29.6$ ($r^2 = 0.91$), all in microequivalents per liter; the 95% confidence intervals for these two regressions do not overlap. PIR, pre-Industrial Revolution (around 1800). Data for SO₄²⁻ and NO₃⁻ in 1940 and 1963-1964 and C_B in 1940 are estimated [for 1940, see (17); C_B was 143 μeq liter⁻¹ in 1963, but anions were not measured]. Values for SO₄²⁻ and NO₃⁻ were estimated from a 1:1 linear extension of the relation at PIR, assuming an ANC of 20 μeq liter⁻¹.

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present using our measurements (18). Concentrations of strong acid anions and Ca^{2+} (and C_0) in stream water likely increased after 1940 because of increasing atmospheric inputs of strong acids. Apparently, acid rain directly or indirectly (19) enhanced the release of Ca^{2+} (and C_0) from forest soil from the mid-1950s until the early 1970s (Figs. 1D, 2, and 3). As the more labile pool of C_0 in soil was depleted, the concentration of C_0 in stream water decreased (1970 to 1994) (Figs. 1D, 2, and 3). Our initial measurements in 1963 probably captured the end of the first phase of rapidly increasing acidification of soil and drainage water and the marked depletion of Ca^{2+} (and C_0) in soil (Figs. 2 and 3).

Increase in streamwater concentration of C_0 during the 1960s corresponds with high but decreasing inputs of Ca^{2+} (and C_0) from atmospheric deposition and large net losses from the soil; declining concentrations of Ca^{2+} (and C_0) in stream water thereafter coincide with smaller atmospheric inputs and decreases in net biomass storage and net soil loss (Fig. 3). Other studies suggest that more than 50% of the Ca^{2+} supply to surface organic soil exchange and vegetation cation pools of forest soils in the northeastern United States may be derived from atmospheric deposition (20).

We estimate that annual bulk precipitation input of Ca^{2+} from 1940 to 1955 was seven times greater than that from 1976 to 1993, and average annual loss from soil from 1955 to 1975 was 1.9 times greater than that from 1976 to 1993 (21). Before the mid-1950s, annual Ca^{2+} depletion from the soil exchange complex was about equal to net biomass storage, and atmospheric deposition plus weathering release nearly

balanced streamwater loss. After the mid-1950s, soil depletion of Ca^{2+} was greater (1.4 times from 1955 to 1975 and 2 times from 1976 to 1993) than net biomass storage, and streamwater loss was larger (2.2 times from 1955 to 1975 and 2.7 times from 1976 to 1993) than bulk precipitation plus weathering release (Fig. 3). Bulk precipitation provided some 29% of the Ca^{2+} required in the ecosystem to balance net biomass storage plus streamwater loss before the mid-1950s; however, from 1955 to 1975, bulk precipitation input decreased to meet only 8% of the total demand, while net soil release increased to 77% (Table 1).

Marked declines in the total Ca^{2+} and Mg^{2+} concentrations of surface organic soil at HBEF have been measured since 1970 (22), and declines have been calculated or anticipated in soil elsewhere in the northeastern United States (19, 20, 23). Appreciable acidification of forest soils by acid rain during the past 50 to 60 years also has been observed in Sweden (24).

Similarly, the Ca^{2+} content of tree rings increased in the 1960s at several locations in the eastern United States. These changes have been attributed to the increased mobilization of C_0 in surface organic soils by acid rain (25). Stemwood calcium has decreased since the 1960s as the pool of exchangeable C_0 in the soil has become depleted (Fig. 1E).

Annual forest biomass accumulation at HBEF has declined unexpectedly to a small rate since 1987 (9), perhaps because available Ca^{2+} became limiting to forest growth as C_0 was depleted from the soil (23). Pinpointing the cause should become a major area of research. Calcium-to-aluminum ratios for soil waters in high-elevation forest stands at HBEF are in a range that may have adverse effects on the growth or nutrition of forest vegetation (26).

After 1963, the average streamwater pH increased slightly at HBEF (pH 4.85 in 1963 to 5.01 in 1993) (Fig. 1C), but the ANC has been consistently negative (currently about $-6 \mu\text{eq liter}^{-1}$). Before the mid-1950s, the ANC in stream water probably was positive. Extrapolating the current

rate of decreases in concentrations of strong acid anions and the corresponding changes in C_0 in stream water at HBEF, we predict that ANC in stream water will become positive again around autumn 1999 (Fig. 2). Nevertheless, even with major reductions in emissions anticipated from the 1990 amendments to the Clean Air Act (27), it is unlikely that the acid-base status of stream water will return to pre-Industrial Revolution levels in the foreseeable future (Fig. 2). Moreover, the large depletion of C_0 from the soil will retard the recovery of the forest ecosystem in response to any decreases in emissions of SO_2 .

Our analysis suggests that forest ecosystems are much more susceptible to continuing atmospheric inputs of strong acids than expected on the basis of trends in sulfur biogeochemistry alone. The long-term hysteresis pattern in streamwater chemistry suggests that ecosystem acidification may be reversible, but recovery is very slow (Fig. 2). A major policy implication of this analysis is that the 1990 amendments to the Clean Air Act will not be adequate to protect surface waters and forest soils of the northeastern United States against further anthropogenic acidification.

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8. ANC is $2[\text{Ca}^{2+}] + 2[\text{Mg}^{2+}] + [\text{Na}^+] + [\text{NH}_4^+] + 3[\text{Al}_3^+] - 2[\text{SO}_4^{2-}] - [\text{NO}_3^-] - [\text{Cl}^-] - [\text{F}^-]$, where the brackets denote concentration in moles per liter; Al_3^+ is the nonhydroxide complexes of aluminum.
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11. The sum of base cations C_0 is $\text{Ca}^{2+} + \text{Mg}^{2+} + \text{Na}^+ + \text{K}^+$ in microequivalents per liter.
12. On the basis of hydrologic considerations, annual values for HBEF are calculated for a water year, 1 June through 31 May (4).
13. Chloride is a small contribution to long-term bulk precipitation (9.0%) and stream water (9.1%) fluxes of strong acid anions at HBEF.
14. See, for example, C. V. Cogbill and G. E. Likens, *Water Resour. Res.* **10**, 1133 (1974).

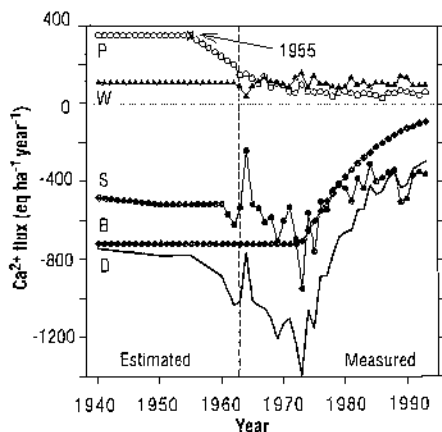


Fig. 3. Annual ecosystem fluxes of calcium for Watershed 6 of the Hubbard Brook Experimental Forest from 1940 to 1963 (estimated) and 1964 to 1994 (measured). Fluxes: P is bulk precipitation input, W is weathering release, S is streamwater loss, B is net biomass storage, and D is net release from the soil exchange complex. Data for P during 1955–1956 are from (15).

Table 1. Percent of ecosystem demand (streamwater loss plus biomass storage) for calcium provided by bulk precipitation (P), weathering release (W), and net release from the soil exchange complex (D).

Period	P (%)	W (%)	D (%)	Total demand (eq ha ⁻¹)
1940–1955	29	9	63	19,610
1955–1975	8	15	77	25,770
1976–1993	12	8	79	12,300
1940–1993	17	10	73	57,680

15. C. E. Junge and R. T. Werby, *J. Meteorol.* **15**, 417 (1958). Samples were collected from July 1955 through June 1956.

16. Estimated Mg^{2+} concentration represents 32% of the 1955–1956 C_0 value. If the Mg^{2+} concentration were zero, which is unrealistic, the C_0 value would be $36.2 \mu\text{eq liter}^{-1}$, or about three times the current value.

17. Sulfate in pre-Industrial Revolution (PIR) (around 1800) stream water was estimated at $20 \mu\text{eq liter}^{-1}$ [T. J. Sullivan *et al.*, *Nature* **345**, 54 (1990); T. J. Sullivan, in *Acidic Deposition and Aquatic Ecosystems*, D. F. Charles, Ed. (Springer-Verlag, New York, 1991)], and NO_3^- was estimated at $5 \mu\text{eq liter}^{-1}$, equaling $25 \mu\text{eq liter}^{-1}$ for $\text{SO}_4^{2-} + \text{NO}_3^-$. PIR values of ANC were probably about $20 \mu\text{eq liter}^{-1}$, resulting in a C_0 estimate of about $45 \mu\text{eq liter}^{-1}$. Initially, in response to increases in emissions of SO_2 and consequent atmospheric deposition of SO_4^{2-} , C_0 concentrations in drainage water probably increased stoichiometrically with increases in SO_4^{2-} . Empirical relations between SO_2 emissions and stream SO_4^{2-} at HBEF suggest stream concentrations of $\text{SO}_4^{2-} + \text{NO}_3^-$ of $96 \mu\text{eq liter}^{-1}$ in 1940. If C_0 increased stoichiometrically with increases in SO_4^{2-} , C_0 and ANC would have been 116 and $20 \mu\text{eq liter}^{-1}$, respectively, in 1940.

18. The weathering rate of Ca^{2+} was estimated from Ca:Na (molar ratio) in bedrock, assuming that all streamwater Na^+ was derived from rock weathering. Calcium release from weathering before 1963 was assumed to be the same as the average from 1963 to 1993, $105 \text{ eq ha}^{-1} \text{ year}^{-1}$. Before 1973, net biomass storage was assumed to be constant ($718 \text{ eq ha}^{-1} \text{ year}^{-1}$) [on the basis of F. H. Bormann and G. E. Likens, *Pattern and Process in a Forested Ecosystem* (Springer-Verlag, New York, 1979)], and after 1973, its value came from a logistic curve fitted to data from 1965 to 1992. Although a logistic curve can be fitted to bulk precipitation input from 1956 to 1994 ($r^2 = 0.87$), we used linear trends and projected the 1955–1956 value ($350 \text{ eq ha}^{-1} \text{ year}^{-1}$) to 1940. Estimated streamwater Ca^{2+} output was based on a relation between SO_2 emissions and concurrent $\text{SO}_4^{2-} + 5 \mu\text{eq liter}^{-1}$ of NO_3^- . Before 1963, the amount of precipitation and streamflow was taken as the average for 1963 to 1993.

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21. The total depletion of Ca^{2+} from labile soil pools during 1940 to 1995 was estimated at $42,200 \text{ eq ha}^{-1}$ (Fig. 3). If Ca^{2+} bulk precipitation concentrations were low from 1940 to 1995, such as currently observed ($3.6 \mu\text{eq liter}^{-1}$), depletion of soil Ca^{2+} would have been $50,400 \text{ eq ha}^{-1}$. In contrast, if Ca^{2+} precipitation concentrations were high from 1940 to 1995, such as observed in 1955–1956 ($25 \mu\text{eq liter}^{-1}$), soil depletion of Ca^{2+} would have been $34,400 \text{ eq ha}^{-1}$. As a result, reductions in atmospheric deposition of Ca^{2+} associated with particulate emission control (5) contributed some 20% of the apparent depletion of soil Ca^{2+} at HBEF.

22. Total Ca^{2+} and Mg^{2+} concentrations in Oe and Oa soil horizons for Watershed 6 of HBEF declined by 30 to 40% between 1970 and 1987 [R. D. Yanai, T. G. Siccama, M. A. Arthur, *Bull. Ecol. Soc. Am.* **74**, 496 (1993)]. Elsewhere in the region, Ca^{2+} in forest floors declined by 22% on average during 1979 to 1994, and Mg^{2+} by 14% [R. D. Yanai, M. A. Arthur, T. G. Siccama, C. A. Federer, *Bull. Ecol. Soc. Am.* **76**, 290 (1995); (19)].

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in Bh and 0.27 in Bs horizon solutions. In high-elevation (730 m) deciduous stands, Ca:Al decreased from 1.21 in Oa to 0.59 in Bh and 0.39 in Bs horizon soil water.

27. We used current NO_3^- plus 50% of the 1980 SO_4^{2-} values to estimate the minimum trajectory of strong acid anion concentrations after 1999 in Fig. 2 (5).

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29. Financial support was provided by the National Science Foundation and the A. W. Mellon Foundation. We thank the various contributions of M. A. Arthur, C. Eagar, C. E. Johnson, C. W. Martin, T. G. Siccama, and R. D. Yanai. The HBEF is operated by the U.S. Department of Agriculture Forest Service, Radnor, Pennsylvania.

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History of Ancient Copper Smelting Pollution During Roman and Medieval Times Recorded in Greenland Ice

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Determination of copper concentrations in Greenland ice dated from seven millennia ago to the present showed values exceeding natural levels, beginning about 2500 years ago. This early large-scale pollution of the atmosphere of the Northern Hemisphere is attributed to emissions from the crude, highly polluting smelting technologies used for copper production during Roman and medieval times, especially in Europe and China. This study opens the way to a quantitative assessment of the history of early metal production, which was instrumental in the development of human cultures during ancient eras.

Most emissions of pollutants to the global atmosphere of our planet occurred after the Industrial Revolution, driven by the combination of unprecedented growth in the population of the Earth and massive technological and economic development. The only exception reported so far is lead emission. Time series recently obtained from Greenland ice (1) and Swedish lake sediments (2) indicate that there was early hemispheric-scale atmospheric pollution from lead during Greek and Roman times. The importance of this early lead pollution remained relatively limited, however; cumulative lead deposition to the Greenland ice cap during the eight centuries of the flourishing of the Greek and Roman civilizations (~400 metric tons) was about an order of magnitude lower than lead deposi-

tion from post-Industrial Revolution fallout [~3200 metric tons (3)]. Here we report evidence that cumulative early atmospheric pollution from copper before the Industrial Revolution was one order of magnitude greater than that from the Industrial Revolution to the present.

We analyzed copper in 23 sections of the 3028.8-m Greenland Ice Core Project (GRIP) deep ice core (4), drilled in central Greenland at Summit ($72^\circ34'N$, $37^\circ37'W$; elevation 3238 m above sea level). Twenty-one sections were selected from ice dated from 2960 to 470 years ago (from depths of 619.3 to 129.3 m) in order to cover the Greek and Roman civilizations, barbaric and medieval times, and the Renaissance. Two sections were chosen from ice dated from 7260 and 7760 years ago (depths of 1230.4 and 1286.5 m, respectively) to assess natural Holocene copper concentrations during times before copper production. We mechanically decontaminated each core section using ultraclean procedures by chiseling successive veneers of ice from the outside to the central part as described in detail elsewhere (5). Copper concentrations were then determined with a precision of $\pm 10\%$ by graphite furnace atomic absorption spectrometry (GFAAS) after preconcentration. Each value was blank-corrected (5). The accuracy of the data depends on the efficiency of the decontamination procedure. This was systematically checked for all sections by investigation of radial changes in copper concentrations from the outside to the center of each individual core section (6). Copper concentrations measured in the central parts of the

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*This paper is dedicated to the memory of C. C. Patterson.

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