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SOME PERSPECTIVES ON LONG-TERM BIOGEOCHEMICAL RESEARCH FROM THE HUBBARD BROOK ECOSYSTEM STUDY

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Abstract. Integrated, long-term, ecological, hydrological, and biogeochemical studies have been done at the Hubbard Brook Experimental Forest since 1963 using the small watershed–ecosystem approach. Some biogeochemical results from these long-term studies, collectively called the Hubbard Brook Ecosystem Study, are described here, including approaches used for managing this large and complicated project. Some major biogeochemical findings of the Hubbard Brook Ecosystem Study are discussed briefly, including acid rain, and some biogeochemical research questions, opportunities, and challenges for the future are identified, including a recent whole-watershed addition of calcium silicate, weathering processes, and long-term trends in stream-water nitrate concentrations.

Key words: acid rain; calcium; chemical budgets; forest disturbance; Hubbard Brook Experimental Forest; net stream retention; nitrogen; pH; watershed–ecosystem approach.

INTRODUCTION

The study of ecosystems as units of nature provides critically important ''windows'' on ecosystem function and change, and on environmental problems (Likens 2001). Insights gained from the use of these windows are especially important currently, as there is increasing demand for scientific information at large scales (landscapes, regions, and the biosphere), for addressing widespread environmental problems.

In 1960, F. Herbert Bormann proposed the use of small watershed–ecosystems for the study of ecosystem function and the connection of the ecosystem to the atmosphere and hydrosphere in a letter to Robert S. Pierce of the USDA Forest Service (Bormann 1996). But, it was not until Herb and I combined our diverse interests and talents that the small-watershed technique became a functional reality. Thus, 40 years ago, joined by Pierce and Noye M. Johnson, we applied the small watershed–ecosystem approach to begin the study of the ecology and biogeochemistry of watershed–ecosystems of the Hubbard Brook Experimental Forest (HBEF) in the White Mountains of New Hampshire. This approach allowed direct measurement of the linkages among the atmospheric, hydrologic, and geologic components of these watershed–ecosystems. It allowed estimates of how the biosphere influenced and was influenced by these relationships (Bormann and Likens 1967: Fig. 1). Answers to important ecological questions about air and water quality, forest growth and sustainability, weathering rates, and ecosystem structure and function in complicated natural landscapes are difficult to obtain. We believed in 1963 that the watershed–ecosystem approach would be useful for tackling such complicated problems.

Our initial approach to the ecosystem conundrum took the form of an analogy. We postulated that we could use the chemistry of stream water draining a watershed similar to the diagnostic approach a physician uses in measuring the chemistry of blood or urine of a human patient. Thus, we needed to determine, simultaneously and quantitatively, all chemical inputs and outputs to the watershed–ecosystem. Such input–output measurements allowed calculation of nutrient budgets (mass balances) for the watershed–ecosystem. Then, combined with experiments and modeling, we were able to address quantitatively, diverse environmental questions at a watershed/landscape scale (e.g., Likens et al. 1970, Bormann and Likens 1979, Rosenberry et al. 1999).

Measurement of inputs to these small, naturally occurring forest ecosystems (tens of hectares in size), provided a measure of how the atmosphere influenced the forest and interlinked stream ecosystems through the input of rain, snow, particles, and gases, and associated chemicals. We quickly realized that the atmosphere was laden with pollutants from distant human activities (e.g., Likens et al. 1972). The measurement of outputs allowed us to determine how water passing through the watershed–ecosystem was altered by the ecosystem and, in turn altered the ecosystem. Analysis of output water, like analysis of blood and urine in humans, became a measure of the ''health'' of the ecosystem, providing insights into some of the complicated, but basic functions of the ecosystem, such as weathering and evapotranspiration (e.g., Likens and Bormann 1995). Since output water was linked to the local and regional hydrosphere, our output measurements provided a means for evaluating the effects of local ecosystem management on regional systems. Out-

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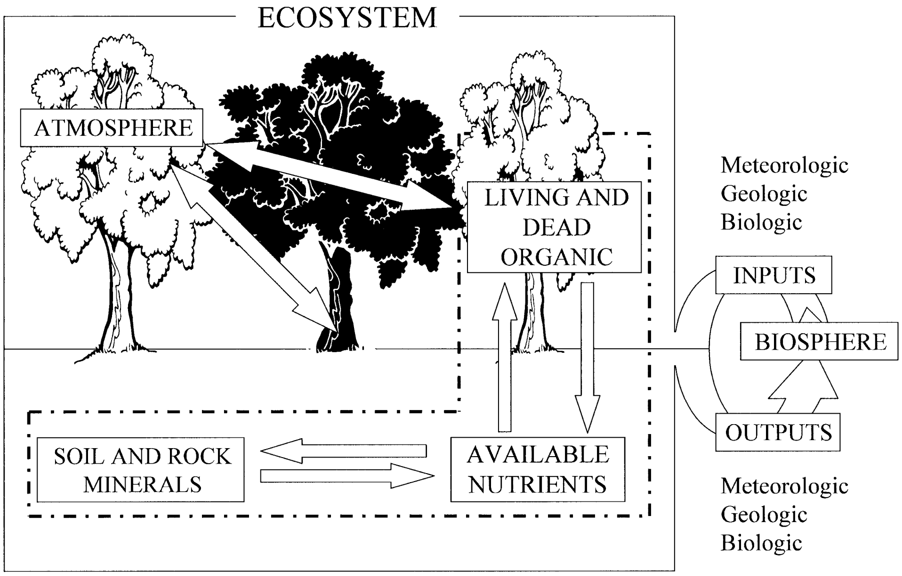

FIG. 1. Nutrient relationships in a terrestrial ecosystem (modified from Bormann and Likens [1967]). Inputs and outputs to the ecosystem are moved by meteorologic, geologic, and biologic vectors (Bormann and Likens 1967, Likens and Bormann 1972). Major sites of accumulation and major exchange pathways within the ecosystem are shown. Nutrients that, because they have no prominent gaseous phase, continually cycle within the boundaries of the ecosystem among the available nutrient, living and dead organic matter, and primary and secondary mineral components, tend to form an intrasystem cycle. Fluxes across the boundaries of an ecosystem link individual ecosystems with the remainder of the biosphere.

puts of gases linked to global atmospheric circulation. Collectively, linking the atmosphere, the hydrosphere, the biosphere, and the implications of these linkages to ecosystem management provided a powerful tool for thinking about local, regional, and global planning and development. The potential of this approach was postulated in our first scientific publication (Bormann and Likens 1967), e.g., ''. . . the rate at which an ion is released by weathering must equal its rate of net loss from the ecosystem plus its rate of net accumulation in the biota and organic debris. . . . Thus, net ionic losses from an undisturbed, relatively stable terrestrial ecosystem are a measure of weathering within the system. . . . Acceleration of losses or, more specifically, the disruption of local cycling patterns by the activities of man could reduce existing 'pools' of an element in local ecosystems, restrict productivity, and consequently limit human population.''

SOME MAJOR FINDINGS OF THE HUBBARD BROOK ECOSYSTEM STUDY

There have been numerous, extremely interesting, if not surprising, discoveries from the long-term research of the Hubbard Brook Ecosystem Study (HBES). A few, directly related to biogeochemistry, are given here:

1) Using the small-watershed approach and measurement techniques developed for the HBES, we established quantitative, input–output, chemical budgets for undisturbed northern hardwood forest ecosystems, showing long-term, net retention (precipitation inputs $>$ stream-water outputs) of H^+, N, Cl, and P and net losses (stream-water outputs $>$ precipitation inputs) of Ca, Mg, Na, K, S, Si, and Al (see Likens and Bormann 1995).

2) Through an experimental disturbance, the deforestation of an entire watershed–ecosystem, an array of ecosystem responses was initiated, primarily the loss of biological regulation of outputs, and with time after disturbance, gradual recovery of biological regulation of outputs (e.g., Likens et al. 1970, Bormann and Likens 1979). The primary effect of deforestation was a severe reduction in evapotranspiration with a shift in evaporative water to runoff, and to increased storm flow. A major finding was that forest cutting not only had a major effect on hydrology, as expected, but also on microbial activity and nutrient output (e.g., Likens et al. 1970, Bormann and Likens 1979). Decomposition and especially nitrification were greatly accelerated with the production of H^+ and NO_3^- that facilitated large losses of nutrients in stream water. Cutting and enforced devegetation also caused an increase of the erodibility of the devegetated system with time. Other natural disturbances such as severe soil freezing or ice storm damage similarly increased NO_3^- loss in stream water from watershed–ecosystems at HBEF (Likens and Bormann 1995, Mitchell et al. 1996, Bernhardt et al. 2003, Houlton et al. 2003).

3) ''Acid rain,'' the popular term for acidified (pH < 5.2) rain, snow, sleet and hail, fog and cloud water, and direct deposition of acidifying gases and particles, was discovered in North America at the HBEF (Likens

et al. 1972). Long-term data from the HBEF provided important ecosystem understanding and information necessary for the passage of the 1990 Clean Air Act Amendments (CAAA).

4) Calcium and other plant nutrients have been markedly leached from the soils of the HBEF by acid deposition (Likens et al. 1996, 1998).

5) Based on extensive and diverse experimental manipulations at the HBEF, we learned that stream ecosystems of the HBEF do not function like ''Teflon pipes.'' Instead, they are active sites of uptake and processing of nutrients and organic matter (e.g., Meyer 1979, Bernhardt et al. 2002, 2003, Hall et al. 2002). Solute pulses added to streams are rapidly attenuated as the solutes move downstream. In-stream retention and processing can significantly reduce the overall net losses from the watershed (see Bernhardt et al. 2003).

LONG-TERM STUDIES

Long-term data have been used to evaluate the biogeochemical response to and recovery from disturbance such as from acid rain, forest cutting, ice storms, and from experimental watershed manipulations. Such long-term records are critical for understanding complicated changes in ecosystem structure and function. These qualitative and quantitative records also provide quantitative information for decision makers wrestling with major environmental issues.

One of the most profound insights from the HBES, especially of experimentally manipulated systems, is that complex legacies play out over long periods. Each disturbance creates a set of conditions or trajectories that impacts the next situation, and thus, the sum total of ecosystem processes is influenced by historical events, each event being overlaid on some previous one. Our long-term ecological and biogeochemical data from the HBES have been invaluable for unraveling such legacy effects, as well as for providing continuity for examination of critical questions, for identifying extreme events, for generating new research questions, and for detecting environmental change. The long-term ecological and biogeochemical record at the HBEF increases in intrinsic value with every year of record added to it. A few examples of these long-term studies include:

1) We discovered that cutting the forest sets in motion an amazing array of biogeochemical changes in ecosystem processes and interactions with concomitant changes in environmental conditions (e.g., Likens et al. 1970, Bormann and Likens 1979). As a result of such experimental manipulations, microbially dominated processes (decomposition, mineralization, and nitrification) were accelerated; ecosystem processes governing the loss of evaporative water (transpiration) were markedly slowed; stream flow was greatly increased as water previously evaporated by the intact forest became liquid water. Initial models of the rate of return to steady-state conditions in these forest ecosystems following clear cutting involved measurement of solutes in stream water exiting the watershed–ecosystem. For ions such as NO_3^-, Ca^{2+}, and K^+, there were large net losses generally peaking in the second year after cutting (e.g., Likens et al. 1970, Bormann and Likens 1979, Martin et al. 2000). Thereafter, stream-water losses declined as the vegetation recovered and net losses of dissolved chemicals returned to near pre-cutting levels at rates unique to each ion. For the purposes of understanding the ecosystem effects of forest cutting and for planning future forest management strategies, these data were clear and sufficient (Likens et al. 1978). Yet, decades after these experimental clear cuts were done within the HBEF, subtle to large differences still can be seen in several stream-water solutes, such as Ca^{2+} (Bailey et al. 2003, Hamburg et al. 2003). Understanding gained from our long-term element mass-balance analyses suggests that the legacy from a 1965–1966 deforestation experiment on Watershed 2 was still affecting ecosystem function in 2003!

2) Declines in emissions of lead, associated with the elimination of leaded fuels in the United States, were correlated with a marked decrease of lead in precipitation and in the forest floor at the HBEF (e.g., Johnson et al. 1995). These data helped confirm the efficacy of regulations against the use of leaded gasoline in the United States.

3) Organic debris dams, naturally formed in streams within a forest landscape, play major functional roles in the ecology and biogeochemistry of stream ecosystems (e.g., Bilby and Likens 1980, Hedin et al. 1988, Steinhart et al. 2000). It was found that 100 years or more are required for organic debris dams in headwater streams to reform following their loss due to disturbance from deforestation (Likens and Bilby 1982, Hedin et al. 1988).

4) Enigmatically, net accumulation of forest biomass has ceased since 1982 at the HBEF (see Likens 2001, Likens et al. 1994, 2002). Is this result a natural response of ecosystem development or some complicated effect of air pollution? Failure of the northern hardwood forest ecosystem to accumulate biomass could have serious implications for the sustainability and harvest of forest landscapes in the northeastern United States. This important question is the subject of intense, ongoing investigation.

Unfortunately, it is not possible here to describe the results of the numerous other long-term, biogeochemical studies from the HBES, including the ecology, biogeochemistry, and hydrology of stream and lake ecosystems (see Likens 1985*a*), experimental ecosystem (''Sand Box'') studies (e.g., Bormann et al. 1993), etc., but one example of our long-term studies is described in more detail in the next section.

ACID RAIN

The primary source of acid rain is the combustion of fossil fuels, which releases sulfur dioxide (SO_2),

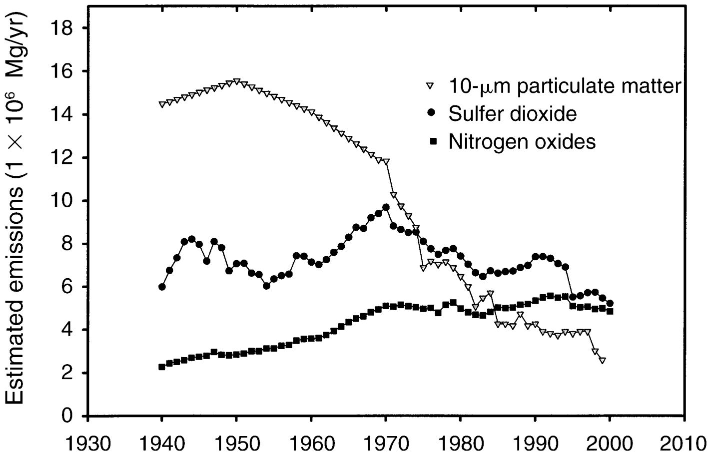

FIG. 2. Long-term trends in emissions of sulfur dioxide, nitrogen oxides, and particulate matter (10 µm in diameter) from the Hubbard Brook Experimental Forest source area (updated from Butler et al. [2001], and Likens et al. [2001]).

nitrogen oxides (NO_x), and acidifying particles to the atmosphere (e.g., Likens et al. 1979, Weathers and Likens 1998). The SO_2 and NO_x may be converted in the atmosphere to sulfuric and nitric acids, and, along with the gases themselves and acidifying particles, are eventually returned to Earth's surface. These inputs acidify some terrestrial and aquatic ecosystems resulting in diverse impacts including the loss of species and accelerated leaching of nutrients, such as Ca^{2+}. Anthropogenic acid rain is a relatively recent environmental problem, but now is spread widely around the world and particularly in eastern North America, northwestern Europe, and eastern Asia. Acid rain represents an experiment at a grand scale being imposed by humans on diverse ecosystems around the world.

As combustion of fossil fuels increased in the United States following the beginning of the Industrial Revolution, emissions of SO_2 and NO_x increased. Recently, atmospheric emissions of SO_2 and small particles have decreased in the United States due to federal regulation (e.g., Likens et al. 2001). In contrast, NO_x emissions, which are largely unregulated, have increased (Fig. 2). We learned from the very first sample of rain we collected at the HBEF in June 1963 that the rain was acid, but it took several years to discover the cause and the nature of its widespread occurrence (Likens et al. 1972, Cogbill and Likens 1974, Likens and Bormann 1974). Sulfuric acid has been the dominant acid in precipitation at the HBEF since 1963, but nitric acid is expected to dominate by 2010–2015 if current trends in emissions continue (Likens and Fallon Lambert 1998). This dramatic change is likely to have significant ecological and biogeochemical consequences on recipient ecosystems.

Undoubtedly, however, the amount and mix of emissions will continue to change in the United States as a result of changing energy demand and pending federal and state actions (e.g., Driscoll et al. 2001). Given the great cost and angst involved with this legislation since the mid 1960s, it is important to measure the legislation's impact on atmospheric deposition and on recipient forest and aquatic ecosystems that have now become highly sensitive to these acidic inputs.

The causes, distribution, and effects of acid deposition have been studied and debated in North America for three decades. Federal regulations to control air pollution in the United States were significantly strengthened and enlarged in 1970 primarily to reduce particulate emissions, but the CAAA in 1990 were the first national legislative initiative, which focused directly on the problem of acid rain. Significant reductions of SO_2 emissions did not occur until 1995 when implementation of Phase I of the CAAA caused a decline in U.S. emissions equivalent to ~40% of the overall reduction targeted by the CAAA. Surprisingly, given the strong correlation between emissions of SO_2 and SO_4^{2-} concentrations in precipitation at HBEF (Fig. 3), this sharp drop in emissions did not produce an unusual decline within the long-term record of precipitation chemistry at HBEF (Likens et al. 2001). Subsequent reductions in SO_2 emissions have been much smaller (Likens et al. 2001).

An extremely important finding from the long-term data at the HBEF was the clarification of the relationship between gaseous emissions of SO_2 and concentrations of SO_4^{2-} in precipitation. This contentious issue had dominated the national debate during the 1980s in the absence of long-term data. We found that both precipitation and stream-water concentrations of SO_4^{2-} are significantly correlated with emissions of SO_2 from the source area upwind of the HBEF (Likens et al. 2002: Fig. 3). Atmospheric deposition of NO_3^- at HBEF also is correlated with NO_x emissions from the source area (Butler et al. 2003). Moreover, there is a strong correlation ($r^2 = 0.76$) between the decrease in stream-water concentrations of SO_4^{2-} observed at the HBEF and the decrease in base cation (Ca^{2+}, Mg^{2+}, Na^-, and K^+) concentrations in stream water (Likens et al. 1996, 1998, 2002). This is an important finding as the base cations are important controls on the acid-neutralizing capacity of the terrestrial and aquatic ecosystems at the HBEF.

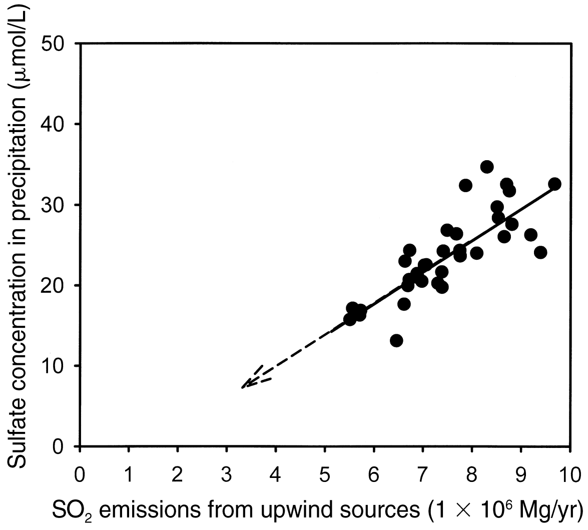

FIG. 3. Annual SO_2 emissions vs. SO_4^{2-} concentrations in bulk precipitation at the Hubbard Brook Experimental Forest (updated from Likens et al. [2001]). Annual SO_2 emissions from the source area (Connecticut, Delaware, Massachusetts, Maryland, New Hampshire, New Jersey, New York, Ohio, Pennsylvania, Virginia, Vermont, and West Virginia) are based on 15-h air-mass, back-trajectory analysis (see Butler et al. 2001).

Our long-term biogeochemical research advanced the knowledge base needed for developing policy, Federal legislation and management related to air pollution. For example:

1) Changes in emissions of SO_2, as a result of Federal legislation, are strongly correlated with changes in SO_4^{2-} concentrations in precipitation and stream water at the HBEF (Likens et al. 1996, 1998, 2001). Thus, reducing emissions of SO_2 will directly reduce inputs of acidifying SO_4^{2-} onto recipient ecosystems.
2) Eighteen years of continuous study were required to verify that the acidity of precipitation had decreased at the HBEF (Likens 1989*a*). The acidity of precipitation decreased from about 90 μeq H^+/L in the mid 1960s to about 55 μeq H^+/L in the late 1980s. However, current values are still about eight times more acid than if the precipitation were not polluted (about 5.2 μeq H^+/L; Fig. 4).
3) Calcium and other plant nutrients have been markedly depleted in the soils of the HBEF as a result of acid deposition (Likens et al. 1996, 1998). As much as one-half of the pool of exchangeable Ca^{2+} in the soil at HBEF has been depleted during the past 50 years by acid deposition (Likens et al. 1996, 1998). These losses may be affecting the biological productivity of the ecosystem. As a result of these losses in soil buffering, forest and aquatic ecosystems have become more sensitive to the degradation from acid deposition than previously thought (Likens et al. 1996).

The HBEF is an important site for monitoring atmospheric pollutants in the northeastern United States because of the long and high-quality record of precipitation chemistry, its location ‘‘downwind’’ of major sources, and lack of local, major sources of pollution.

Recently, a whole watershed manipulation was initiated to test experimentally some of the long-term effects of acid deposition on the ecosystems at the HBEF (see Peters et al. 2004). A natural calcium silicate mineral (Wollastonite), mined in the Adirondack Mountains of New York State, was pulverized, pelletized, and then added to Watershed 1 at the HBEF by helicopter in October 1999. An amount of calcium estimated to have been depleted from the ecosystem during the past 50 years was added in this manipulation. It is planned to study the effect of this experimental manipulation on stream and soil chemistry, tree growth, animal populations, microbial activity, and other components of the watershed–ecosystem over the next 50 years.

MANAGEMENT OF THE HBES

Long-term continuity of a complex study, such as the HBES, involves much more than science alone. Several management features and goals were fundamental to sustaining the productivity and integrity of the HBES over 40 years; (1) developing at the outset and continuing to use a conceptual biogeochemical model (Fig. 1) for guiding research and ecosystem analysis; (2) nurturing a strong incentive within our team to understand the whole system (the ecosystem) rather than a reductionist approach of focusing exclusively on the components (parts) of the system; (3) integrating results and preparing synthesis volumes as rapidly as possible; (4) nurturing the concept of long-term studies even though it was often difficult to maintain uninterrupted funding; (5) enticing outstanding colleagues from a variety of disciplines to join our scientific team, often without financial incentives; (6) maintaining a

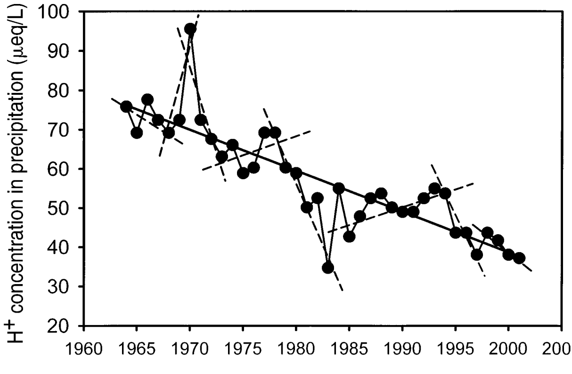

FIG. 4. Short- and long-term trends in hydrogen ion concentration in precipitation at the Hubbard Brook Experimental Forest (updated and modified from Likens [1989*b*]).

TABLE 1. Some major challenges for biogeochemical studies in the future (modified from Likens [2004]).

1) What are the specific effects and relationships of the increasing size of the human population on the biogeochemical flux and cycling of elements, and the effects of forcing functions often incongruent in space and time?
2) What controls fluxes of N and P to and from natural and human-dominated (cities, agricultural) ecosystems?
3) What is the quantitative role of stream/river ecosystems in watershed retention of nutrients and how does it change over time?
4) What is and what controls C sequestration in diverse ecosystems (e.g., forest, ocean, lakes, wetlands) on variable temporal and spatial scales?
5) What controls weathering rates, and what are the sources and fates of the weathered products, including nutrient loss in terrestrial ecosystems?
6) What are the quantitative interrelationships among hydrology, ecology, and biogeochemistry?
7) How can a better synoptic understanding of the biogeochemical flux, cycling, and interaction of elements among air, land, and water (including ocean) systems be achieved?
8) What are the critical linkages and feedbacks among major nutrient and toxic element fluxes and cycles?

small, focused, and dedicated team of researchers who spent much time in residence, interacting together *at* the HBEF; and (7) developing analytical procedures that were neither changed nor replaced without first overlapping and comparing results from the ''long-term method'' with those from a proposed new method. This procedure helped to avoid ''artifacts'' in the long-term, biogeochemical data. Any such changes were carefully documented (Buso et al. 2000).

Entire-watershed, experimental manipulations have been a powerful analytical tool of the HBES. In the words of a colleague, William Lewis, Jr. (2002), ''Watershed manipulation now is a standard part of the biogeochemist's repertoire, but in the 1960s it must have seemed radically intrusive and perhaps even a bit pushy. . . (Experimental) manipulation, as we now know, vastly accelerates the pace of discovery, and that was one of the secrets of success for what became known as the Hubbard Brook Ecosystem Study.'' The ability to do such large-scale, experimental manipulations on a long-term basis, with adjacent watersheds for reference (see Likens 1985*b*), indeed, was one of the scientific joys and successes of the HBES. We are grateful to the USDA Forest Service, particularly to Robert S. Pierce, for acceptance and support in this regard.

Computer simulation models were developed by our colleagues and applied as important research and predictive tools for analyzing long-term hydrologic and biogeochemical trends and processes (e.g., Federer and Lash 1978, Aber et al. 2002, Gbondo-Tugbawa et al. 2002).

These scientific management approaches to the HBES were central to any successes. Additional detail regarding our operating and management philosophy for the HBES is given in the prefaces to our first two synthesis volumes (Likens et al. 1977, Bormann and Likens 1979).

THE FUTURE OF THE HUBBARD BROOK ECOSYSTEM STUDY

It is extremely difficult, if not impossible, to predict the future, particularly in these uncertain times. Nevertheless, the long-term chemical record combined with the long-term hydrologic record and stable research infrastructure provided by the USDA Forest Service at the HBEF have served, and are likely to serve well into the future, as a magnet for research and for researchers of the HBES. There is no lack of exciting questions, challenges, and research opportunities for studies of ecosystem structure and function, and biogeochemistry (e.g., see Likens 2001, 2004). The HBEF has been an NSF-funded LTER site since 1988, an EPA-funded NDDN (National Dry Deposition Network) site since 1988 (changed to CASTNet [Clean Air Status and Trends Network] in 1990) and a USGS site for hydrologic studies since 1980 and a site for characterizing Ground-water Flow and Chemical Transport in Fractured Rock since 1990. Some difficult biogeochemical problems that we have wrestled with for 40 years will continue well into the future, e.g., dynamics of the N cycle (fixation, denitrification, and ecosystem retention). For example, annual, volume-weighted NO_3^- concentrations in stream water are currently the lowest in 40 years at the HBEF (e.g., Likens 2001). Will these low values continue in spite of negligible forest biomass accumulation? How will the weathering process be evaluated, including the relative importance of plagioclase, apatite, and oxalate as sources of Ca^{2+} within watershed ecosystems (e.g., Likens et al. 1998, Blum et al. 2002, Bailey et al. 2003)? What are the overall impacts of acid deposition on vegetation, soils, and aquatic ecosystems? What is the quantitative role of stream ecosystems in nutrient processing and watershed retention? Many new questions will also emerge (Table 1).

Dozens of senior scientists, students, and technicians do research at the HBEF every year. It seems likely that they will do so for at least another 40 years, and will be driven by persistent questions that remain vital to science and society, and by new questions that are generated from the long-term data and from the effects of new perturbations imposed on the biogeochemistry of this most remarkable and scientifically valuable valley.

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