Integrated Acidification Study (ILWAS): A Mechanistic Ecosystem Analysis [and Discussion]

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Integrated acidification study (ILWAS): a mechanistic ecosystem analysis

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An integrated, interdisciplinary, intensive study of three forested watersheds was started in 1977 to quantify the relationship between the deposition of atmospheric acids and surface water acidity. Results indicate the importance of using an integrated ecosystem perspective to assess the vulnerability of surface waters to acidification and the value of analysing relative flowpath contributions to understanding surface water alkalinity levels and dynamics. Important ecosystem properties affecting surface water acidity are soil depth, soil mineralogy and stage of forest development.

ILWAS objective

Lake surveys have indicated that the acidity of some surface waters has increased during this century in regions of Norway, Sweden, Canada, and the United States. This acidification has been linked qualitatively with the deposition of atmospheric acids. The objective of the Integrated Lake–Watershed Acidification Study (ILWAS) is to establish the quantitative nature of the link; that is, to derive a general theoretical framework for characterizing rates of change in the acidity of surface waters as a function of change in levels of atmospheric acid deposition. This framework should take into account watershed biogeochemical processes that produce and consume acidity.

To evaluate the effectiveness of various pollution control and acidification mitigation strategies, it is necessary to characterize quantitatively with time how non-acidic lakes would react to continuing acidic deposition at current levels and how all lakes would respond to changed levels of acidic deposition and various neutralization schemes. The vehicle being developed by ILWAS for making these characterizations is a mathematical model that simulates, in an integrative manner, the multiple biogeochemical processes controlling lake–watershed acidity. The model is formulated by using mechanistic biogeochemical concepts to be able to apply the results of ILWAS to geographical areas other than the Adirondacks, where the field work has taken place. In essence, the model can be considered a means by which the general mechanistic theory of surface water acidification developed by ILWAS is defined, the theory being a complex set of relationships involving multiple state variables.

Study description

ILWAS is a large, intensive, integrated, interdisciplinary, multi-institutional study of three forested watersheds (Goldstein *et al.* 1980). Approximately a dozen principal investigators from as many as nine institutions made up the core study team. The study was started in the autumn of 1977. Field data collection was completed in December 1981. Final reports are in preparation; however, the data set, which consists of over 600000 measurements, and the model, are so robust that meaningful analyses of both will probably continue for many more years.

The conceptual basis of the study derives from the recognition that precipitation follows various pathways through a terrestrial system before it reaches a lake. Biogeochemical processes acting in series and in parallel, on a landscape level, produce or consume acids and release chemicals that shift the pH and alkalinity of natural waters. Scientific investigation of a single process is not sufficient for understanding the chemical behaviour of rain as it becomes lake water. The results of one process may be modified by others to yield results that are counterintuitive and conflicting. An integrated approach is needed.

Based on a conceptual model (Chen *et al.* 1979), the study watersheds have been divided into a network of compartments: atmosphere, canopy, snowpack, soil system, bogs, streams and lake. Data were collected for each compartment to evaluate its acid producing and consuming processes and to calibrate and verify the ILWAS mathematical model that simulates the quality and quantity of water moving through the watershed.

Field investigations were conducted to measure atmospheric inputs, watershed characteristics (for example, distribution of plant communities and depth of soil) and the chemistry of ground and surface waters. Frequency of sampling varied. Precipitation was sampled at each precipitation event. Soil water was sampled biweekly, while the lake outlets were sampled twice weekly and groundwater monthly. Laboratory experiments were conducted to investigate processes such as mineral weathering, anion adsorption, decomposition and nitrification.

Key aspects of ILWAS are: (i) well-defined objective and scope; (ii) simultaneous measurement of watershed inputs, outputs and state variables; (iii) simultaneous study using identical experimental design and methods on multiple watersheds that appeared very similar but whose responses to similar atmospheric inputs were dramatically different; (iv) team approach that emphasized periodic meetings of all principal investigators and 'intensive' sampling periods when all principal investigators went into the field to study major system transients (for example, flush of acidity accompanying snowmelt); (v) intensive collection of field data for a four year period; (vi) emphasis on processes, mechanisms and integration; (vii) emphasis on quantification; (viii) concurrent execution of modelling and data collection programmes allowing feedback between the programmes and modification of both during the project period; (ix) application of model as an integration tool to test consistency among data subsets and between the model's conceptual basis and the data; and (x) periodic review by peer scientists. While some of the above aspects are unique for an ecosystem analysis, the fundamental uniqueness of ILWAS is its incorporation of all of the above aspects.

The fundamental assumption on which the design of ILWAS was based is that a lake's vulnerability to acidification by atmospheric deposition can only be understood in the context of the biogeochemistry and hydrology of its entire catchment. All ILWAS results to date have supported this assumption. In a short paper such as this, we are not able to present fully the general theory of surface water acidification developed by ILWAS with analyses of all major factors affecting surface water acidity and their multiple interactions. Our objective in this paper is to use the ILWAS model to examine how several environmental factors affect lake acidity and while doing so illustrate the necessity of using an ecosystem perspective to analyse surface water acidification.

SITES

The study sites consist of three forested watersheds located within 30 km of each other in the Adirondack Park region of New York State. The bedrock of the area is granitic and the tree communities are coniferous and deciduous. The coniferous communities are dominated by spruce (*Picea rubens*) and fir (*Abies balsamea*) and the deciduous communities by beech (*Fagus grandifola*), birch (*Betula* spp.) and maple (*Acer* spp.). Soils are developed mainly on glacial till and their upper layers are strongly acidic. Quartz and K-feldspar are the most abundant minerals in the till. The most abundant reactive mineral, from the perspective of weathering to produce alkalinity, appears to be hornblende (April & Newton 1983). However, because of their great abundance, feldspar minerals, although less reactive than hornblende, probably significantly contribute to alkalinity production (R. H. April, personal communication). Annual precipitation in the area tends to be between 100 and 140 cm. The median rain event pH is about 4.2 (Johannes *et al.* 1981). Detailed information about the geology, hydrology, soils, vegetation and water chemistry of the watersheds is given in E.P.R.I. reports (1981 and 1983).

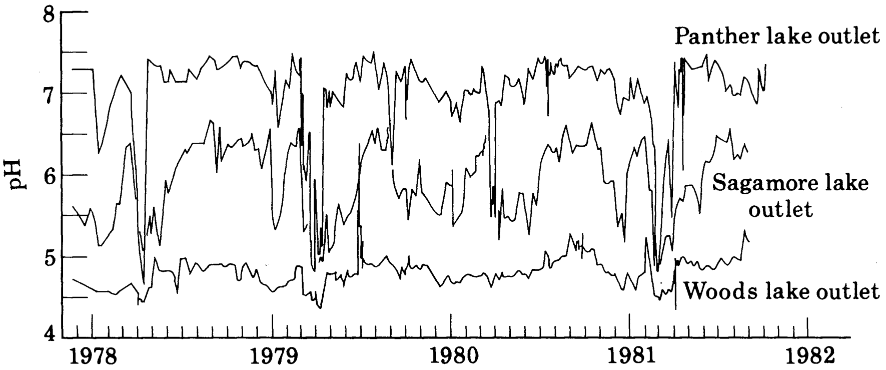

FIGURE 1. Air equilibrated outlet pH of Panther, Sagamore and Woods lakes over the duration of the ILWAS field programme.

Although each watershed receives about the same amount of precipitation of nearly identical quality, each has a lake with a different pH level and different pH dynamics (figure 1). Of the three lakes, Woods is considered acidic (typical outlet pH between 4.5 and 5.0), and Panther is neutral (typical outlet pH near 7). Sagamore Lake has a much larger watershed (49.0 km^2 compared to 2.1 km^2 and 1.2 km^2 for Woods and Panther, respectively) with more spatially heterogeneous biogeochemical characteristics, that results in more variable year-round pH dynamics with an outlet pH typically between that of Woods and Panther.

The lakes tend to be well mixed vertically because of wind action (mean depth varies from 3.5 m in Panther to 4.0 in Woods to 8.8 in Sagamore), with well defined thermal stratification only occurring under ice cover (Hendrey *et al.* 1980). Outlet chemistry therefore reflects the chemistry of the entire lake, except during the period of thermal stratification when it reflects most closely the chemical characteristics of the water at the lake surface. During spring snowmelt, Panther Lake's outlet undergoes a rapid transient increase in acidity (figure 1). Acidification does not occur over the entire depth of the lake but only at the surface (Hendrey *et al.* 1980). Soon afterward, as the surface waters warm, the lake mixes and returns rapidly, as does the outlet, to a neutral pH.

Mathematical model

Within the study, the role of the model is to organize the measurement of lake–watershed acidification processes into an integrated theoretical framework. To accomplish this, the model simulates the major acid producing and consuming process, routes the flow of water, and calculates the quality and quantity of water in each component of the lake–watershed system.

The model is divided into two modules: hydrological and chemical. The hydrological module (Chen *et al.* 1982) routes water from tree top to lake outlet. It simulates the hydrological processes of canopy interception; throughfall; snow accumulation and melting; soil freezing and thawing; lake freezing and thawing; evapotranspiration and soil, stream, bog and lake hydraulics.

The chemical module simulates biogeochemical processes occurring in the canopy (Chen *et al.* 1983*b*), snowpack, soil, streams, bogs and lake that determine the concentrations of the chemical constituents in the water in each of these compartments. Simulated processes include dry deposition, foliar exudation, snow leaching, litterfall, nitrification, oxidation, decomposition, nutrient uptake by terrestrial and aquatic plants, root respiration, mineral weathering, cation exchange, anion adsorption, Al dissolution–precipitation, and chemical equilibration in water.

Simulated chemical species and parameters include: SO_2, NO_x, H^+, NH_4^+, Al (monomeric), Ca^{2+}, Mg^{2+}, K^+, Na^+, SO_4^{2-}, NO_3^-, Cl^-, F^-, $H_{3-n}(PO_4)^{n-}$, alkalinity, total inorganic carbon (t.i.c.), H_4SiO_4, and organic acid analogues.

The simulation time step is a day. Time dependent model input includes daily precipitation, maximum and minimum daily temperatures, monthly averaged precipitation quality, and monthly averaged ambient air quality. Other input consists of constants, parameters and functions that characterize the biological, chemical and physical properties of the watershed.

To apply the model, a watershed is divided into horizontally homogeneous drainage subcatchments, stream segments and a lake. A bog can be represented by a stream segment. The connectivity of water flow among these elements is defined. Each subcatchment is segmented vertically into compartments representing canopy, snowpack and soil layers. The segmentation of the soil will be determined by how its properties vary with depth and considerations concerning the numerical stability of the simulations. Each soil layer is characterized by such properties as thickness, field capacity, saturation water capacity, permeability, bulk density, adsorbed cation and anion densities, cation exchange capacity, mineralogical composition, and percentage organic matter.

The lake is also segmented vertically for simulating heat and chemical fluxes. The lake layers have a constant thickness but different horizontal dimensions. Layers can be added (or subtracted) as the water surface rises (or falls). The top layer has a variable thickness, which may be smaller than the thickness of the other layers. A detailed documentation of the entire model has been published by E.P.R.I. (Chen *et al.* 1983*a*).

Flowpath

Precipitation falling into a watershed may travel different pathways before reaching a lake. Some precipitation falls directly into the lake and streams. The rest is intercepted by the forest canopy. That water which is not returned to the atmosphere by evapotranspiration, can drain through the soil to any of a number of depths and then flow laterally to streams or the lake. The actual flowpath taken at any given time will be a function of environmental conditions and physical soil properties. Different flowpaths will expose a parcel of water to different strength sources and sinks of alkalinity. Data taken by Cronan (1983) demonstrate how the pH of soils and soil water in Panther and Woods watersheds increase with depth. It is significant to note that pH values for soil solution from the same soil layers in the two watersheds are similar.

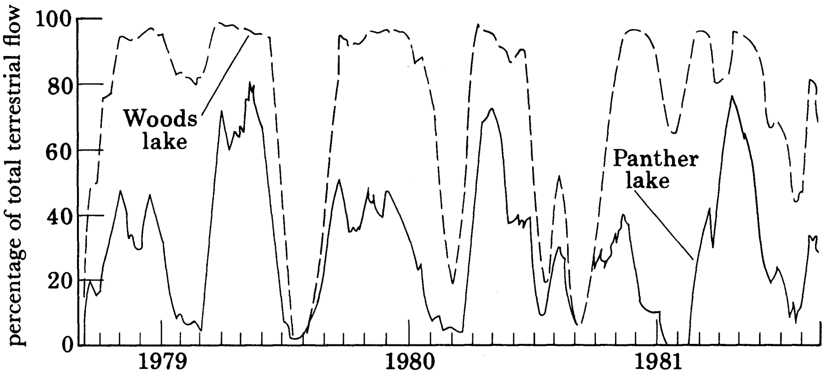

FIGURE 2. Simulated surface plus interflow as a percentage of simulated total terrestrial inflow to Panther and Woods lakes. Percentages are based on 30 d moving averages of flow.

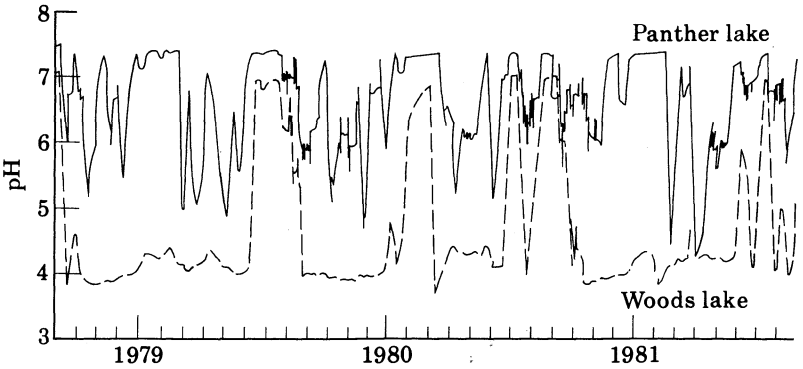

FIGURE 3. Mixing cup pH of the sum of all terrestrial inflows to Panther and Woods lakes. Calculations are based on assumption that water is in chemical equilibrium with CO_2 at three times atmospheric CO_2 pressure. Curves represent 10 d moving averages.

Even through a lake itself water can take different flowpaths depending on the degree to which the lake is stratified. Waters following different flowpaths will have different chemical characteristics. The chemistry of a stream or lake will be a function of the relative contribution of different flowpaths to the waterbody. It is important to recognize that the relative contribution of different flowpaths to a given waterbody is not time invariant and can change seasonally or over longer time periods with changing environmental conditions.

We have used the ILWAS model to illustrate the influence of flowpath, by calculating (figure 2) the ratio of simulated litter and organic layer flow (interflow) to total inflow for both Woods and Panther Lake. We have also calculated (figure 3) the pH of the inflow to the lakes. The inflow pH is calculated by mixing the water from all of the flowpaths and allowing it to equilibrate at three times atmospheric CO_2 pressure, which is a representative of CO_2 partial pressure for the lakes. When comparing the two figures, note that figure 2 plots a 30 d moving average and figure 3 a 10 d moving average. This results in smoother curves in figure 2. The moving averages were selected to optimize the clarity of the illustrations.

The first thing to note in figure 2 is the considerably greater relative contribution that interflow makes to Woods Lake. This is consistent with the different measured pH levels of the outlets of the two lakes (figure 1) and corresponds to the inflow pH simulations (figure 3). Next note the peaks in relative interflow contribution that occur in Panther watershed during spring. This is consistent with the transient pulses of acidity measured in Panther Lake outlet during snowmelt and corresponds to the simulated inflow pH pattern. What is interesting and surprising are the peaks in relative interflow contribution that occur in Panther in the autumn, since with the exception of September 1978 no major spikes of increased acidity are observed in the autumn in the outlet of Panther Lake. The inflow pH simulation, however, corresponds to the flow pattern and shows autumn spikes of increased acidity. The explanation for the dissimilarity between the patterns of simulated inflow pH and observed outlet pH in the autumn is that the lake is well mixed which results in a dilution and hence dampening out of the inflow spikes. If it is assumed that the September 1978 spike in the outlet is real, it is not clear what environmental conditions could lead to its occurrence. The sampling station is somewhat downstream of the outlet weir. Perhaps the spike represents an acidification of the outlet stream and not the lake outlet or perhaps there was something unique about the environmental conditions at that particular time that allowed the acidity to cross the lake without being diluted by mixing. It should be noted that although a Panther-type lake would not be very vulnerable to acidification episodes other than during the snowmelt period, this lack of vulnerability would not necessarily apply to small streams in a Panther-type watershed. Autumn acidification events have been frequently observed in other geographical areas, such as the Tovdal catchment in southern Norway (Overrein *et al.* 1980).

In figure 2, we note sharp declines in the relative contribution of interflow to Woods Lake during summer. This corresponds to sharp increases in simulated inflow pH. Again because of lake mixing we would not expect to observe these fluctuations at the lake outlet (hydraulic retention time in Woods during summer is about eight months), and do not with the single exception of the summer of 1979. There is no ready explanation for this single event. Analogous to autumn acidifications in a Panther-type watershed, although a Woods-type lake would not be subject to large increases in pH during the summer, streams on a Woods-type watershed could. Large summer increases in alkalinity have been frequently observed in other geographical areas, for example, the Tovdal catchment (Overrein *et al.* 1980) and have also been observed in Adirondack streams during the dry summer of 1983 (C. N. Schofield, personal communication).

If the simulated inflow pH curves are compared to the measured outlet pH curves, we note that, even though the inflow curves are 10 d moving averages, the outlet curves are considerably smoother. The process of physical mixing in the lake acts as a strong buffer and dampens fluctuations in inflow chemical concentrations.

Soil depth

Soil depth is a major factor producing the different hydrologies for Woods and Panther watersheds. As stated previously, soils on both watersheds are developed mainly on glacial till overlaying granitic bedrock. Chemical, physical and mineralogical soil properties are similar. One difference between the two soils is that Woods contains a thin layer of aeolian silt overlaying the till (Newton & April 1982). The silt has lower permeability than the rest of the soil; however, since the silt layer is discontinuous, it is not clear that it significantly reduces drainage in Woods watershed relative to Panther. (A sensitivity analysis of the model has shown that a 25% areal coverage with aeolian silt has no effect. Larger percentage coverages have not been simulated.)

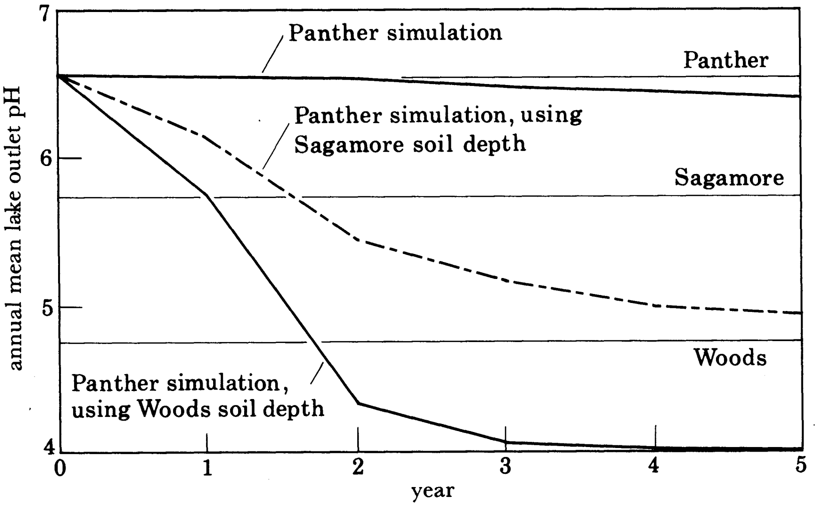

FIGURE 4. Annual mean lake outlet *in situ* pH for simulations of Panther Lake watershed using soil depths of Panther, Sagamore and Woods watersheds. Horizontal lines are the measured annual mean outlet *in situ* pHs of the three lakes.

The most dramatic difference in the soils between the two watersheds is depth. Based on seismic refraction measurements, Newton & April (1982) calculate the average depth of Woods to be 2.3 m and the average depth of Panther to be 24.5 m. The average depth of Sagamore is in between.

The ILWAS model has been used to simulate the effect of soil depth on outlet pH. In the first case (figure 4), the ILWAS model calibrated for Panther watershed has been run for three sets of mineral soil thicknesses, corresponding to the thickness of Panther, Sagamore, and Woods. Before time 0, all three simulated systems are identical to Panther, then at time 0 soil depth is altered. Several years are required for the altered systems to approach a new equilibrium outlet pH. In the second case (figure 5), the model calibrated for Woods has been run for mineral soil thicknesses corresponding to Woods, Sagamore and Panther watersheds. As with the previous set of simulations, alteration of depth occurs at time 0. Both figures compare the simulations to the observed annual mean outlet (*in situ*) pH of the three lakes.

Changing mineral soil thickness changes the volume of weatherable minerals, exchangeable base cations and anion adsorption sites, and the relative contributions of different flowpaths to lake inflow. Increasing mineral soil thickness with no other changes in soil physical properties decreases the ratio of litter and organic layer flow to total inflow. As discussed in the previous section, this produces increased lake pH and alkalinity.

While soil depth appears to be a good single key indicator for differentiating between the acidic status of Panther and Woods Lakes, it is essential to recognize that soil depth would not necessarily be a good single key indicator in more general situations. For instance, for catchments where net transfer of water is from lake to groundwater or where a large volume percentage of groundwater flows by the lake and does not interact with it, the relative contribution of deep soil water flow to the lake could be relatively independent of soil depth. We have observed these situations in seepage lakes in the Adirondacks and in northwest Wisconsin that are sites in a follow-on study to ILWAS.

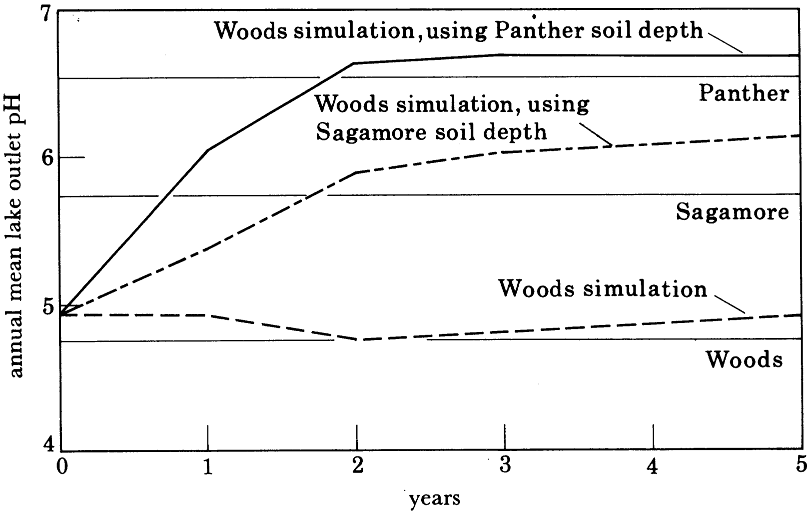

FIGURE 5. Annual mean lake outlet *in situ* pH for simulations of Woods Lake watershed using soil depths of Woods, Sagamore and Panther watersheds. Horizontal lines are the measured annual mean outlet *in situ* pHs of the three lakes.

SOIL MINERALOGY

In earlier sections, the deep mineral soil has been identified as a major reservoir of alkalinity. The degree to which this reservoir is tapped depends on the flow routing of water. The two principal processes that produce alkalinity in the mineral soil are mineral weathering and cation exchange. In this section, using the ILWAS model, we explore the relation between these two processes and alkalinity production.

To avoid complexities in interpretation resulting from horizontal spatial non-uniformity of watershed characteristics and to decrease computer time needed for multiyear runs, simulations were conducted on a hypothetical single catchment lake–watershed whose characteristics are similar to the ILWAS sites and whose lake is non-acid but not as alkaline as Panther (summer outlet alkalinity of about 150 μeq l^{-1} compared to 200 for Panther).

Simulations were conducted for two alterations of the watershed. In the first, all hornblende was removed from the soil. In the second, cation exchange capacity (c.e.c.) was set to zero in all layers but the organic. Cation exchange capacity is left in the top soil layer (organic layer) to minimize the chance of forest productivity becoming nutrient limited. Not surprisingly, the simulations reveal that although cation exchange summed over the entire depth of soil is a net source of alkalinity, the exchanger in the uppermost soil layer is a sink for alkalinity because of its low base saturation.

Simulation results (figure 6) showed that over a period of 30 years, the removal of hornblende led to a drop of only a small percentage in total system net annual alkalinity production (annual lake outflow of alkalinity minus total annual atmospheric input of alkalinity) relative to the unaltered (base) case, while the removal of c.e.c. led to a decline of 35%, despite increased weathering of minerals. The reason for the small effect resulting from hornblende removal is that cation exchange compensated for the alkalinity production lost through the removal of hornblende. It should be noted, though, that the base saturation existing on the exchanger is the result of a long term (thousands of years) equilibration with mineral weathering products. Without this weathering, the pool of adsorbed cations, which compensates for the simulated removal of hornblende, would not be as large.

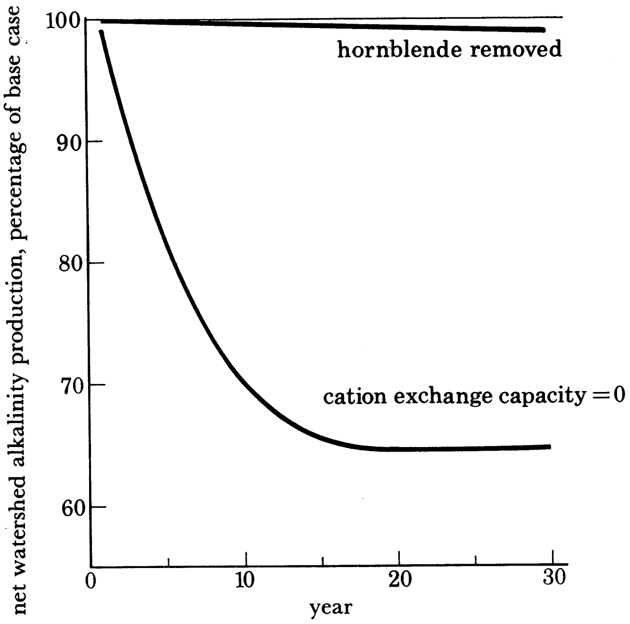

FIGURE 6. Simulation of responses of a hypothetical lake–watershed system to: (*a*) removal of hornblende, and (*b*) setting cation exchange capacity to zero in all layers except the organic. Responses are measured in terms of net annual alkalinity production (annual alkalinity lake outflow – annual total atmospheric alkalinity input) for the altered systems as a percentage of the same flux for the unaltered system.

Figure 7 illustrates the lake outlet alkalinity in year 30 for both the base case and the case where c.e.c. is set equal to zero. Note that the difference between the two cases is small ($\approx 20\ \mu\text{eq l}^{-1}$) at the peak of snowmelt relative to the rest of the year ($\approx 120\ \mu\text{eq l}^{-1}$). This is because at snowmelt most of the water travelling through the terrestrial system into the lake is shallow flow.

This simple exercise emphasizes further the need to place the subject of surface water alkalinity in an ecosystem context and understand the interrelationships among ecosystem sources and sinks of alkalinity. The results illustrate how the contribution of a given source or sink (in this case cation exchange) can be controlled by other sources and sinks, and that in response to a change in one source or sink (in this case hornblende weathering), it is possible for a system to adjust so that surface water alkalinity is practically unchanged. Of course, the

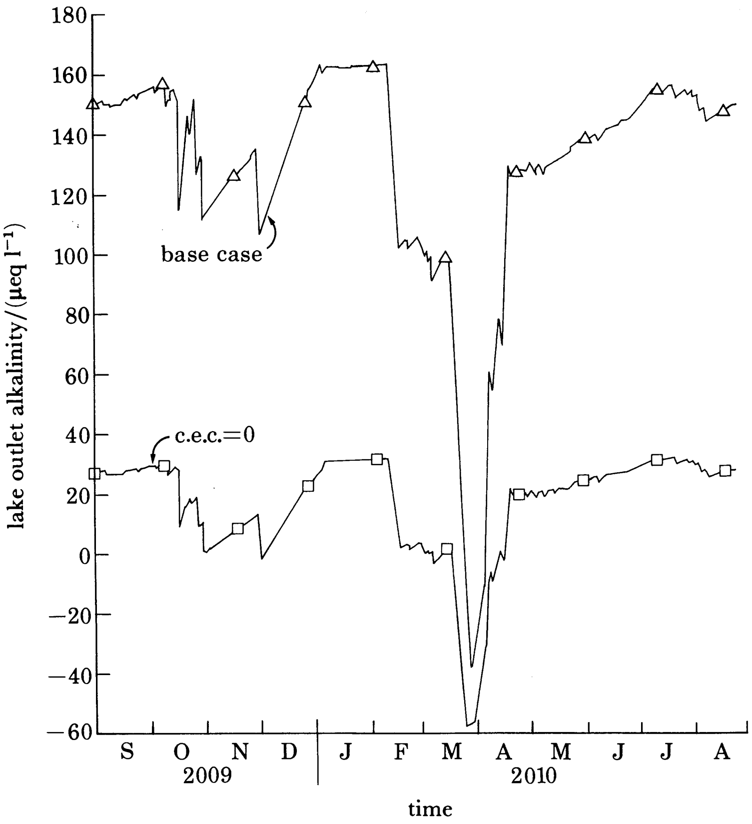

FIGURE 7. Simulations of lake outlet alkalinity for a hypothetical lake–watershed for case where c.e.c. is to zero in all soil layers but the organic and unaltered (base) case. Simulations are for 30th year after the alteration of c.e.c.

nature and magnitude of any compensation depend on lake–watershed and climatic characteristics and the nature of the alteration. The value of a mathematical model to study how the properties of an ecosystem and its environment constrain ecosystem dynamic behaviour has been discussed by Goldstein (1977).

TEMPERATURE

In addition to lake–watershed characteristics, climatic factors also affect the alkalinity of surface waters. Long term climatic trends and multiyear climatic fluctuations can potentially produce major effects.

In this section, we briefly examine some effects of atmospheric temperature. Figure 8 illustrates simulations of the outlet pH of Panther Lake for measured atmospheric temperatures (base case) and with measured temperatures shifted ±2 °C. (The reason that the three curves have different initial pH values in the illustration is that the simulations have already been running for a year before 1 September 1979.)

For the simulated warmer climate, the acidic pulse at snowmelt occurs earlier than the base case, while for the colder climate it occurs later. Alteration of the time of occurrence of snowmelt could alter its potential effect on fish populations, for example, by changing the occurrence of the pulse relative to spawning. On the other hand, temperature changes may alter the time of occurrence of critical life stage events for the fish in the same way that it alters the time of occurrence of the snowmelt.

One might expect that there would be a greater accumulation of snow for lower temperatures, which would result in the minimum pH of the outlet plus snowmelt decreasing as the climate gets colder. Although the simulated depth of the snowpack is greater for colder temperatures (maximum depth of approximately 23 cm of water for the −2 °C case compared to 12 cm for the +2 °C case), the minimum pH does not decrease. The width of the alkalinity depression during snowmelt, however, does increase with colder temperatures. It does not appear possible to generalize about minimum pH, which might be significant with respect to potential effects on aquatic biota, since the same temperature simulations run for another watershed resulted in decreasing minimum pH with colder temperatures.

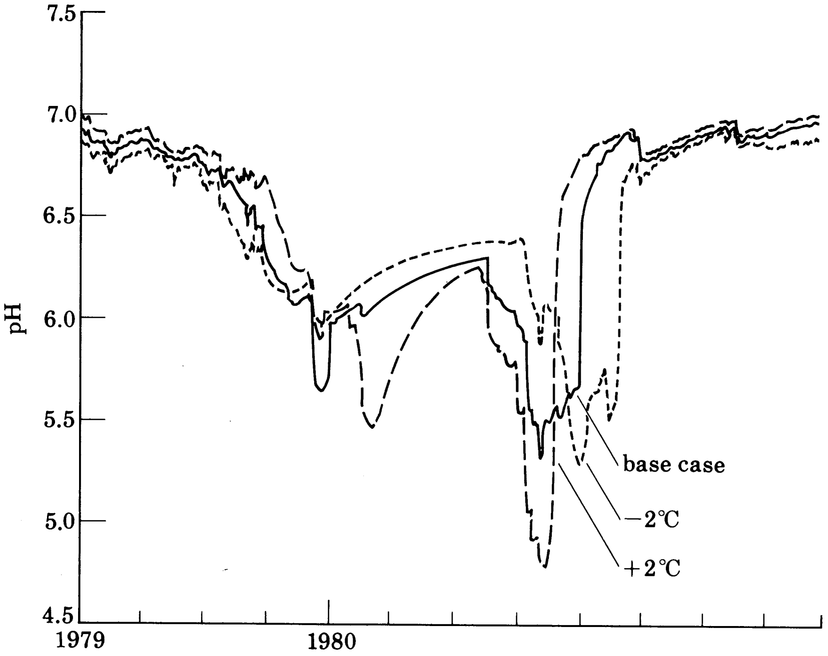

FIGURE 8. Simulations of pH of Panther Lake outlet for measured atmospheric temperatures (base case) and measured temperatures shifted ±2 °C.

There is a simulated midwinter snowmelt in December. Although it appears that the melt is considerably greater for the base case than the +2 °C case, this is not so. In fact, the depressions in alkalinity are similar. The depressions are different on the pH scale because of the initial alkalinity before the melt for each case relative to its pH–alkalinity relationship. The base case starts at a lower alkalinity which puts it on a steeper part of the pH–alkalinity curve, therefore it incurs a more dramatic pH depression for an equivalent alkalinity depression. A separate rise in temperature that occurs in January produces a melting only for the +2 °C case.

The simulations indicate that a cooler climate produces a lower pH during the summer. One might suppose that a cooler climate would produce wetter soils as a result of less evapotranspiration and that this could affect the relative contribution of flowpaths and thus lake alkalinity. However, we checked simulated alkalinity inflow to the lake during summer and noted no significant differences in simulations. Hence, we conclude that the difference in outflow pH during summer is a residual effect of the different snowmelt characteristics for the different climates.

It is important to recognize that the magnitude and nature of the simulated effects discussed above will vary with lake–watershed characteristics and other climatic features. We are in a sense examining a system response (outlet pH) in a highly multidimensional space whose axes consist of all the lake–watershed and climatic factors that govern the response. By varying only one factor and holding all others constant, we are taking a partial derivative in a small region of the entire space. In other regions, the derivative can be dramatically different.

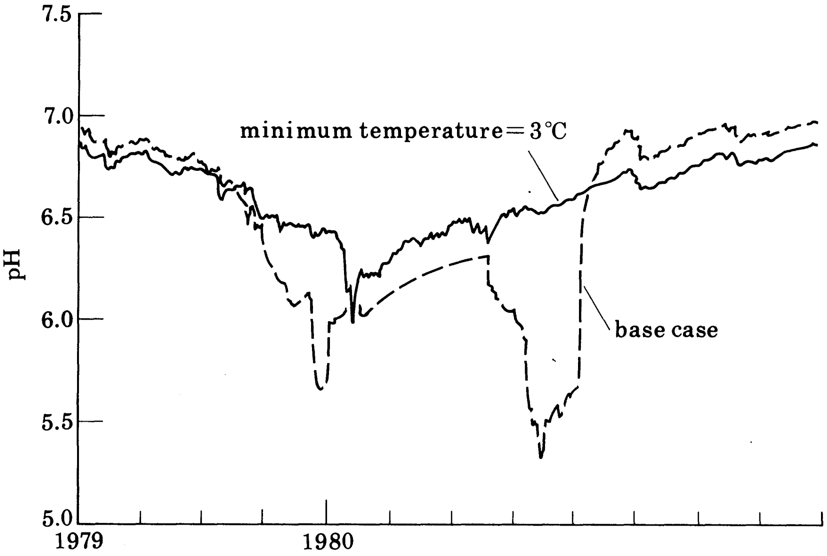

FIGURE 9. Simulations of pH of Panther Lake outlet for measured atmospheric temperatures (base case) and for case where measured temperatures below 3 °C are set at 3 °C.

It is also noted that for simulating climatic effects it would be attractive to develop a stochastic model for generating meteorological conditions. This model would be based on long-term measurements. The climate could then be altered by changing annual or seasonal means or higher moments within the model.

Figure 9, illustrates simulations of outlet pH of Panther Lake for the base case and for the case where atmospheric temperature is not allowed to fall below 3 °C. On days when temperature is below 3 °C, the temperature is set to 3 °C. In addition to converting snow events to rain events, this temperature alteration prevents lake ice formation and minimizes, if not totally does away with, thermal stratification in the lake. As previously discussed, without stratification it is highly improbable that a major pH depression will occur in Panther. Hence the simulated temperature alteration does not allow the effect of the absence of snow to be distinguished from the effect of removing lake stratification. This illustrates how different lake–watershed phenomena are closely integrated and the difficulty associated with trying to separate them. (This observation holds for both the real system and the model.) Snow could be prevented by making the model enter all precipitation events as rain regardless of atmospheric temperature. In such a case, the lake surface would freeze and typical winter stratification would occur but the amount of soil freezing would be altered. Hence the resulting outlet pH would have to be interpreted with respect to both changes in snowpack and frozen condition of the soil. It should be pointed out that changing soil temperature also alters nitrification and decomposition rates.

The clipped temperature simulation (figure 9) results in almost total attenuation of the winter pH depression in the outlet and disappearance of all transient acidic pulses. However, there still is a decline in pH (and alkalinity) in the winter and there is a small residual decrease relative to the base case in the summer.

It is ironic that when this simulation was first run in the autumn of 1982 as a 'what if' scenario, it was assumed to have the same relevance to the Adirondacks as the response to the question 'what if Napoleon had won the battle of Waterloo?' has to modern France. No one could recall a winter in the Adirondacks when there was not some snowpack accumulation. But amazingly, during the winter of 1982–83, no snowpack accumulated. C. Schofield (personal communication) measured Panther Lake outlet pH from the beginning of March until the end of April, which includes the period of icemelt in early April, and did not record an air equilibrated pH below seven (see figure 1 to compare with four previous winters). Unlike the simulation, ice cover and stratification were present. It is unfortunate that a large storm event did not occur before icemelt because it would have been valuable to have seen if an acidic pulse occurred. It will be informative to run the ILWAS model with 1982–83 winter meterological data and compare the outlet simulation to Schofield's data.

Ecosystem perspective

Climatic and lake–watershed characteristics interact in multiple ways to determine surface water chemistry. Many attempts have been made to find a simple single factor to characterize a lake's vulnerability to acidification, for example, bedrock geology, surface-water alkalinity, sulphate concentration in precipitation compared to calcium and magnesium concentration in lake water, vegetation type, soil thickness, soil cation exchange capacity. While one or more of these may be adequate over a limited geographical area where lake–watersheds only differ with respect to a few properties, it appears that in general nature is too diverse for this search to ever be successful. However, it is straightforward to put the subject of surface water acidification into an ecosystem context as was done in ILWAS and analyse in this context the relation between atmospheric deposition and surface water acidity for any given lake–watershed or region.

The concept of relative contribution of flowpaths appears to be a very powerful analytical tool. Of course it should be remembered that flowpath is an integrating concept (not a single factor) and is a function of lake–watershed characteristics and environmental conditions. Also relative contributions of different flowpaths can change both seasonally and with longer term climatic fluctuations or trends. Analyses of hydrographs and short-term water budgets are approaches to calculating the relative distribution of terrestrial inflow between shallow and deep flow.

The necessity of taking an ecosystem perspective is further illustrated by the average annual alkalinity budget calculated for Woods and Panther Lake watersheds for the period January 1978 to December 1981 (table 1). Atmospheric input is calculated using wet deposition measurements and dry deposition simulated by the ILWAS model. Lake–watershed (system) output is calculated using measured volumetric outflow and chemical concentrations of the lake outlets. Forest growth (or net biomass uptake) is estimated based on net system retention of nitrogen (atmospheric input–system output). It is assumed all net retention of nitrogen goes into forest growth. A biomass stoichiometry that defines the proportion of all other elements to nitrogen is assumed. From these proportions and total nitrogen retention, the amounts of all other anions and cations going into forest growth can be calculated. The alkalinity contribution of nitrate and ammonium to forest growth is based on the net system retention of each ion calculated separately. Hornblende weathering is based on an assumed weathering stoichiometry and the calculated net system release of either silicon or chloride. Sulphate adsorption is calculated by subtracting the sulphate that goes into forest growth from the net system retention of sulphate. Net other weathering and ion exchange is calcuated by differing net watershed production and forest growth, hornblende weathering, and sulphate adsorption. Normalization of all fluxes is based on the terrestrial areas of the basins.

TABLE 1. AVERAGE ANNUAL ALKALINITY BUDGET

(January 1978–December 1981)

flux/(equivalents per hectare)	Panther watershed	Woods watershed	error estimate, %
total atmospheric input	−1125	−1090	±20
outflow	1035	−85	±10
net watershed production	2160	1005	
forest growth (net biomass uptake)	−785	−710	±30
hornblende weathering	485	90	±30†, ±60‡
sulphate adsorption	140	100	±20
net ion exchange and other weathering	2320	1525	±20

† Panther. ‡ Woods.

The budget clearly illustrates the importance of forest growth and soil mineralogy in understanding surface water alkalinity. It is important to keep in mind that both relative and absolute contributions of the different soil processes to the net watershed production of alkalinity is very much determined by the flow routing of water moving through the system.

CURRENT RESEARCH DIRECTION

In 1982, the Regional Integrated Lake–Watershed Study (RILWAS), a follow-on study to ILWAS, was started. Two objectives or RILWAS are to test and increase the robustness of the ILWAS model by applying it to areas outside of the Adirondacks and to develop and test a methodology to assess the vulnerability of an entire region (in contrast to a few individual lakes) to acidification by atmospheric acids.

The first objective is being addressed in both northern Wisconsin and the southern Appalachians. In northwestern Wisconsin, we are studying two forested lake–watersheds whose climates, lakes, hydrologies, soils and vegetation are very different from the three original Adirondack lake–watersheds. The Wisconsin lakes are seepage lakes. That is, they have no surface inlets or outflows and thus their hydrologies are predominately influenced by local and perhaps regional groundwater systems. Soils are developed on sandy glacial outwash and are much thicker (of the order of 100 m) than the Adirondacks. The bedrock is Precambrian sandstone. The dominant vegetation consists of stands of jack pine (*Pinus banksiana*) and northern pin oak (*Quercus ellipsoidalis*).

The ILWAS model is also being applied to a watershed in the southern Appalachians in western North Carolina. Southern Appalachian watersheds have a number of unique hydrological, soil and climatic features. The principal surface waters of interest in this region are streams and reservoirs in contrast to the small lakes of Wisconsin and the Adirondacks. Because the southern Appalachian region has never been glaciated, its soils, which are millions of years old, are much older than Wisconsin or Adirondacks soils, which are about 12000 years old. The absence of the formation of a snowpack in the south should permit better resolution of winter watershed sources of nitric acid production than is possible in the north where the snowpack contains considerable nitrogen.

Policy makers and the general public are not so much concerned with the fate of individual lakes as they are with the fate of an entire lake region, hence the second RILWAS objective is to develop a regional assessment methodology that has greater validity than regional one-time surveys of surface water chemistry, but is not as complex, time-consuming, and costly as applying the very intensive ILWAS approach to every lake in a region. This part of the study is being carried out in the Adirondack Park area, where 20 study watersheds have been selected to represent the variety of environmental situations that exist in the region. Watersheds have been selected to include a variety of mineralogies, bedrock types, hydrologies, and fisheries.

In addition to the two objectives described above, RILWAS will attempt to relate lake fisheries to watershed characteristics and biogeochemistries. This part of the study focuses on a network of interconnected lakes with different chemistries and with watersheds having different characteristics. The distribution of fish species through this network will be compared to the lake chemistries and lake–watershed characteristics.

Many ideas in this manuscript arose from stimulating interactions with other ILWAS participants and ILWAS reviewers. Included in this group are: Martin Alexander and Carl Schofield, Cornell University; Nicholas Clesceri, Elmar Altwicker, and Arland Johannes, Rensselear Polytechnic Institute; Richard April, Colgate University; Christopher Cronan, University of Maine; James Galloway, University of Virginia; George Hendrey, Brookhaven National Laboratory; Robert Newton, Smith College; Jake Peters and Dave Troutman, U.S. Geological Survey; David Grigal, University of Minnesota; and Robert Wetzel, Michigan State University.

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Discussion

J. R. Kramer (*Department of Geology, McMaster University, Hamilton, Ontario, Canada*). I refer to the spring snowmelt and pH depression. In 1983, eastern North America had a snowless winter. Yet I understand that a spring pH depression was measured and demonstrated in Wisconsin. Has Dr Goldstein data to run his model for this time period? If so what were the results?

R. A. Goldstein. As discussed in greater detail in the paper, there was no snow accumulation in the Adirondacks during the 1982–83 winter and no pH depression was observed in the outlet of Panther Lake during March and April 1983. However, if while the lake was thermally stratified a large amount of precipitation fell during a relatively short time period, then it is possible that a depression could have occurred. The depression results primarily from an alteration of the relative routing of flow in the catchment. This can occur in response to a number of situations, of which the melting of a snowpack is one.

D. J. A. Brown (*C.E.R.L. Freshwater Biology Unit, Ratcliffe-on-Soar Power Station, Nottingham NG*11 0*EE, U.K.*) Dr Goldstein has described the model forecasts for such factors as changes in temperature or changes in the chemical properties of the soil, but did not describe what the model would predict should happen to the chemistry of the lakes in response to a change in precipitation acidity. Is he prepared to elaborate?

R. A. Goldstein. In general, surface water responses to changes in precipitation acidity could be expected to be highly diverse. The response for a given catchment would depend on its biogeochemical characteristics and climate. The response for a region would most likely be best defined in a probabilistic context that considered spatial distributions of important lake–watershed and climatic factors.

We have not as yet used the model to conduct a thorough analysis of surface water response to changes in deposition acidity. With respect to the Woods and Panther systems, some exercises that could be considered preliminary analyses have been performed. Both the annual average outlet pH and the spring pH depression of Panther Lake appear to be relatively insensitive to changes in deposition acidity. Annual average pH of Panther outlet continues to be neutral in response to both halving and doubling current sulphate deposition. Within the mineral soil, the large volume of exchangeable bases and the high percentage of base saturation of the exchange sites produces a high buffering potential. The snowmelt surge of acidity in the Panther system appears to be primarily a response to the alteration of the relative contributions of different flowpaths to the inflow and outflow of the lake and not strongly dependent on the acidity of the snow.

The Woods system as a result of a smaller buffering potential than Panther appears to be more sensitive. A simulation in which sulphate deposition was reduced to 25% of current levels resulted in an increase, after several years, of outlet pH of about one half unit.

P. F. Chester (*C.E.G.B. Central Electricity Research Laboratories, Kelvin Avenue, Leatherhead, Surrey KT22 7SE, U.K.*). Rosenqvist argues that exchange of base cations for hydrogen ions in the humic layers of soil is an important source of acidity in surface water. One route by which such cations could reach the humic layers is through 'return flow' from mineral layers as described by Dr Bache in his paper. Does the ILWAS model include any term representing the passage of Ca^{2+} or Mg^{2+} by any mechanism from mineral to humic horizons?

R. A. Goldstein. One pathway for moving cations from mineral to organic layers is through the vegetation by means of root uptake, leaf exudation and litterfall. The ILWAS model simulates this movement.

A second pathway is directly through the soil by means of the possible upward movement of soil water if the water potential is lower in the organic than in the mineral soil layers. The ILWAS model does not simulate this phenomenon. On an annual basis, how much water actually follows this pathway? Some years ago, J. B. Mankin and I developed a model (called PROSPER) of atmosphere–plant–soil water flow that simulates explicitly the soil water potential gradient and thus can simulate upward as well as downward flow in the soil (Swift *et al.* 1975). This model can be applied to different ecosystems to judge the importance of the upward flow pathway. Rosenqvist considers this pathway important in the boreal systems that he studies. I do not feel that in general this pathway is significant in temperate forests.

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