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Alkalinity Generation in Snowmelt and Rain Runoff During Short Distance Flow Over Rock

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Abstract

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High-elevation ecosystems in the western United States typically have patchy, discontinuous areas of surficial soils surrounded by large areas of rock outcrop, talus, and scree. Snowmelt and precipitation that percolate through soil increase in alkalinity, principally by increasing base cation concentration through cation exchange, and by decreasing acid anion concentration by adsorption or uptake of sulfate and nitrate. While it is widely believed that changes in chemistry during runoff over rock outcrop and through saprolite contribute to increases in alkalinity, there have been few studies that document the magnitude of this change. In the Wind River Mountains, WY, snowmelt and rain samples increased in alkalinity approximately 35 meq L^{-1} during 15 to 50 meter transport over rock, lichens, and thin pockets of saprolite and soil. Alkalinity increases were principally due to increased base cation concentration. Nitrate concentrations in snow melt decreased from approximately 8 meq L^{-1} to <1 meq L^{-1}; however, nitrate increased following rock transport of rainfall. There were significantly increasing trends in alkalinity generation with increased path length; however, the relationship with path length was not strong.

Keywords: Acid deposition, alkalinity, acid neutralizing capacity, alpine ecosystems.

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Aquatic ecosystems in Western North America are generally not thought to be as sensitive to acid deposition as aquatic ecosystems in the Northeastern United States, Eastern Canada, and Europe. This is due in part to lower emission densities of acid pollutants, but also because soils in the West are generally well buffered. However, some forested montane areas, and particularly higher elevation alpine and subalpine ecosystems located on noncarbonate lithologies are considered to be highly sensitive to acid deposition. This consideration is based on the fact that such areas contain relatively patchy, thin soils, and that chemical weathering rates and accompanying base release are slow because of low temperature and short residence time for water in contact with rock and saprolite. Data from a large sample of Western United States alpine lakes support the contention that these systems are, in fact, weakly buffered (Eilers and others 1987; Landers and others 1987), although chronic acidification has never been documented in the Rocky Mountains (Campbell and others 1995). Aquatic ecosystems are most vulnerable in basins with shallow, coarse-textured soils formed from rock containing minerals that weather slowly, and without appreciable base release (for example, quartzites).

While there have been many studies describing mechanisms and models to predict soil processing of acid deposition, there are few data available describing the acid neutralizing capacity (ANC) generated when precipitation flows over rock outcrop, talus, or scree. Many of the same processes that generate ANC in soils occur during acidic drainage over rock or percolation through saprolite; however, the amount of ANC generation is likely to be less because of higher water velocity, decreased residence time, and lower specific surface of rock and coarse-textured saprolite compared to soil. In addition, pool sizes that contribute to base exchange and anion adsorption are much less per unit area of land when comparing rock to soil.

Large areas of alpine and subalpine land are managed by the U.S. Department of Agriculture, Forest Service as Wilderness Areas in the Western United States. As a Federal land management agency, the Forest Service has responsibility under the Clean Air Act Amendments of 1977 to prevent significant deterioration of air quality (the PSD program). Under the PSD program, managers must identify and describe air quality related values (AQRVs) in Class I Wilderness Areas, and define limits of acceptable change (LAC) for the identified values. While AQRVs and LACs vary from Forest Service Region to Region, lakes and streams in alpine ecosystems are typically identified as one example of an AQRV susceptible to change from acid deposition, and LACs generally include some level of change in water alkalinity. LACs for alkalinity are usually dependent on perceived vulnerability of the water body, and projected changes due to increased deposition are model based. Acidification models like MAGIC (Cosby and others 1985a,b), once calibrated for a lake or stream, can be used as a screening procedure for estimated response to changes in acid deposition.

The MAGIC model is a lumped parameter model that includes simulation of changes in water chemistry during percolation through one or two soil compartments. Soil processes simulated include cation exchange, sulfate adsorption, nutrient uptake, pH dependence of weathering, and $Al(OH)_3$ dissolution. The model is calibrated for four lakes in the Bridger Wilderness in Wyoming. The current calibrations include two soil compartments: rock outcrop and a composite of the common soils in the area (Clayton and others 1991a). The model is calibrated by matching simulated lake chemistry to observed lake chemistry data when provided with precipitation, hydrological, and soil parameters. It is possible to calibrate without distinguishing between rock and soil processing of water by providing a set of soil parameters that results in "correct" lake chemistry. However, this approach is undesirable because it is likely to lead to incorrect forecasts under scenarios of changing deposition chemistry. This research characterizes snowmelt and rainwater chemistry change during short-distance transport over rock to provide a guide for rock and soil parameter selection.

Methods

Site Description and Sampling

Snow, rain, and runoff samples were taken in the vicinity of Hobbs Lake in the Bridger Wilderness, Bridger-Teton National Forest (Lat. 43°02'23"N; Long. 109°40'00"W), WY. The Bridger-Teton Forest has collected rain and snow samples at this location as part of their air quality monitoring program since 1986. Five sampling locations at an elevation of 3,100 m were selected to collect snowmelt and rainwater at the base of an essentially vertical rock cliff. The cliff faces generally south. However, snowmelt from the five locations was from southwest-facing portions of the cliff. The rock is a high-grade Archean migmatite or granitic veined gneiss, according to Worl and others (1984), containing quartz, potassium feldspar, plagioclase, epidote, and biotite. Maximum flow path length varied with cliff topography among the five sites, and ranged from 15 to 50 m. Flow paths at each site were classified by length as either bare rock, or soil/vegetation. The soil/vegetation class was either predominantly moss and lichens growing on rock, or fine material (gravel and sand sized) that had accumulated to a depth of a few centimeters on a small bench position on the cliff face. This area is representative and typical of the kinds of areas classified and mapped by soil scientists as rock outcrop, talus, scree, or some other nonsoil classification.

During spring snowmelt sampling, three 500 mL samples of snowmelt dripping from cornices were collected at each of three locations in the vicinity of the cliff (total of nine samples). In addition, four 500 mL samples were collected at the base of each of the rock sites (total of 20 samples). During rainfall sampling, the same protocol was used for flow over rock, but one sample of rain was collected using a Hubbard-Brook (Likens and others 1977) rainfall collector. Snowmelt samples were collected on May 18, 1993, and May 31, 1995. These collections were early in the snowmelt season; however, it is not known whether they might represent an early melt “solute or acid pulse” (Johannessen and Henriksen 1978). A single rain collection was sampled on September 29, 1994. Samples were returned to the Pinedale Ranger District office within 5 hours of collection, and pH and specific conductance read on one sub-sample from each rock site location and each snow collection site, or the single rain sample. Three rock samples from each site and three snowmelt subsamples, or one subsample of rain were filtered through 0.45 μm membrane filters, frozen, and sent to the Boise Forestry Sciences Laboratory for chemical analysis. The remaining rock sample and snowmelt subsamples were not filtered, and were sent to the Rocky Mountain Forest and Range Experiment Station in Fort Collins for chemical analysis.

The Boise laboratory analyzed Na and K by flame emission spectrophotometry, Ca and Mg by flame atomic absorption spectrophotometry, and Cl, SO_4, NO_3, and PO_4 by ion chromatography (IC). Samples collected in 1995 were analyzed for NO_3 by Cd-reduction (Wood and others 1967) as well as IC because the 1993 snowmelt samples that had run over rock were at the limits of our detection using IC. All samples sent to Fort Collins had Gran titrations run to determine alkalinities.

Data Analysis

All precipitation samples were tested for charge balance agreement to assure analytical quality, based on the calculation:

$$[(\Sigma \text{ Cations} - \Sigma \text{ Anions}) / (\Sigma \text{ Cations} + \Sigma \text{ Anions})] \times 100 \qquad (1)$$

where Cations or Anions is the equivalent concentration of positively and negatively charged species. Rock flow samples consistently have larger anion deficits than snowmelt or rain samples, presumably due to organic anions that were not accounted for during analysis. Reuss and others (1993) allowed up to 20 percent cation excess in stream samples to account for dissociated organic acids before rejecting a sample, and all of our rock flow samples satisfied that criterion.

Data were analyzed to test for differences attributable to sample locations on the rock face, and to test for differences between snowmelt versus over-rock flow chemistry. Tests for difference among the five rock sample locations for all collections was done by GLM-ANOVA. The Tukey-Kramer multiple comparison test was run on rocksites that tested significantly different (probability of a larger F <0.05) to compare means at $\alpha = 0.05$. Concentration differences between snowmelt and rock runoff were tested with a two-sample t-test, $\alpha = 0.05$, with $n = 5$ for rock samples and $n = 3$ for snowmelt samples.

Results

Precipitation Chemistry

Snow and rain chemistry from the three sampled events was generally typical of longer term data collected in the area. Table 1 compares chemistry from this study with National Atmospheric Deposition Program (NADP) wet deposition mean values sampled from 1986 to 1994 at four locations near the Wind River Range (Baron 1996), and data from the Hobbs Lake bulk collector site located within 1 km of our sampling site. Mean calcium concentrations for samples

Table 1—Comparison of snowmelt and rain chemistry samples from this study (n = 16) with volume-weighted mean annual values from four nearby NADP wet deposition sites (1986-1994), and arithmetic mean annual values from the Hobbs Lake bulk collector (wet plus dry), sampled from 1992-1994.

	This study		NADP		Hobbs Lake	
Element	**mean**	**s**	**mean**	**range**	**mean**	**range**
	$\mu eq\ L^{-1}$					
H	6.82	2.52	5.6	1.3	11.8	7.4
Na	3.87	2.74	3.75	3.7	4.5	1.2
K	0.75	0.65	0.83	0.8	1.4	1.3
Ca	7.83	2.36	13.0	3.0	17.4	8.1
Mg	1.92	0.64	2.90	1.6	3.6	1.0
Cl	3.18	1.35	3.73	2.3	3.9	2.4
NO_3	8.85	2.02	12.18	4.5	12.4	3.1
SO_4	10.95	2.38	14.42	3.8	13.1	2.9
ANC	–7.52	3.36	–9.85	4.8	–4.7	10.7

from this study were approximately half the mean concentrations measured at NADP sites and the bulk collector samples. NO_3 and SO_4 values are also lower than reported values from the NADP and bulk sample locations. Fourteen of our 15 precipitation samples were snow samples, and the lower Ca concentration may result from eolian dust sources being covered with snow during some or most of the winter months. The single rain sample was collected in a container that was opened during the storm and therefore not subject to accumulations of windblown dry particulates. However, the NADP samples are theoretically not collecting dry deposition.

Changes Due to Flow Over Rock

Flow over rock results in distinct changes to the chemistry of precipitation including: increases in base cation concentration, decreases in NO_3 concentration, increases in pH, and increases in alkalinity. These data are presented in table 2 for all rock sites combined for the three sample dates. With the exception of Na, individual base cation concentrations increase significantly during flow over rock from snowmelt. The magnitude of cation increase is larger for the rain sample collection date; however, the significance could not be tested because of the single rain sample. The

Table 2—Concentration of chemical species in snowmelt, rain, and runoff following rock transport in the vicinity of Hobbs Lake for 1993, 1994 and 1995 events. For snow samples in 1993 and 1995, n = 3; the rain collection in 1994 is a single sample; flow over rock samples for all 3 years, n = 5.

	1993				1994				1995			
Chem.	**snow**		**rock**		**rain**		**rock**		**snow**		**rock**	
species	**mean**	**s**	**mean**	**s**	**value**	**s**	**mean**	**s**	**mean**	**s**	**mean**	**s**
						$\mu eq\ L^{-1}$						
H	3.85*	0.40	0.72	0.25	6.76	—	0.99	0.41	8.81*	0.50	1.13	1.25
Na	5.92	1.39	7.95	3.58	0.40	—	15.07	3.61	2.90	1.53	4.45	1.62
K	0.69*	0.37	3.75	1.94	0.60	—	9.68	2.90	0.81*	0.76	5.38	4.01
Ca	9.38*	2.41	39.6	13.1	2.74	—	63.6	19.7	7.36*	1.50	29.2	13.1
Mg	1.81*	0.65	11.5	4.98	0.31	—	16.6	4.19	2.17*	0.39	9.87	4.50
C_B	17.8*	4.21	62.8	22.9	4.05	—	105	29.3	13.2*	4.03	48.9	21.6
Cl	3.54	0.94	2.53	1.45	0.86	—	18.9	1.75	3.20	0.49	4.52	2.21
NO_3	8.00	2.77	<0.5	—	2.71	—	11.73	11.9	8.08*	0.54	0.83	0.70
SO_4	11.2	4.03	11.8	5.13	8.28	—	33.8	4.61	11.1*	1.23	6.90	1.96
C_A	22.8*	7.64	14.3	6.44	11.9	—	60.7	21.6	22.3*	2.01	11.4	3.01
Alk	–2.95*	2.33	35.1	11.5	–8.10	—	29.9	9.33	–12.4*	2.31	25.5	12.7
ANC	–4.97*	3.58	48.5	18.8	–7.8	—	44.2	18.9	–9.19*	2.36	38.7	20.5

* Means marked with an asterisk are significantly different than rock mean values.

sum of the base cations (C_B) increased 2.5 to 3 times for snow collections, and nearly 20 times for the rain collection (fig. 1).

While base cation concentrations consistently increase with over rock transport, acid anions responded individually and inconsistently following transport over rock. Chloride concentrations in snow did not change significantly. However, Cl in rain increased more than 20 times following rock flow. Nitrate concentrations in snowmelt averaged approximately 8 μeq L^{-1}, and decreased to our detection limits by IC analysis (< 0.5 μeq L^{-1}) following transport over rock (table 2). The 1995 samples analyzed by the more sensitive cadmium reduction method averaged 8.86 μeq L^{-1} before rock flow (8.08 μeq L^{-1} by IC), and 0.83 μeq L^{-1} after. In contrast, NO_3 , like Cl, increased during over rock transport for the rainfall sample. Sulfate concentrations in snowmelt did not change in 1993 following rock transport, but decreased from approximately 11 to 7 μeq L^{-1} in 1995. Sulfate concentration during the rain event increased by over three times. This combination of anion changes resulted in the sum of the acid anions (C_A) decreasing significantly during rock flow for both snow events, but increasing for the rain event (fig. 2).

Alkalinity concentrations in rain and snow ranged from –12 to –3 μeq L^{-1}, and increased 37 to 38 μeq L^{-1} during over rock transport (fig. 3; table 2). ANC, calculated from C_B–C_A, was similar to alkalinity concentrations for precipitation samples, but ANC values

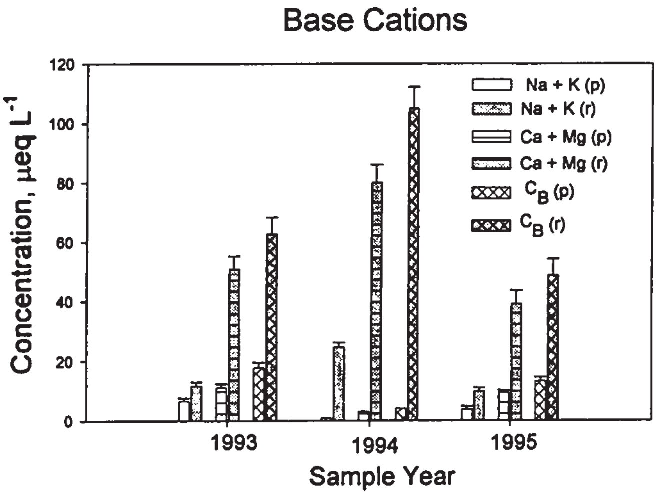

Figure 1—Concentrations of base cations in snowmelt, rain, and runoff over rock for sampling years 1993, 1994, and 1995. C_B is the sum of the base cations. Error bars are one standard error of the mean. Snowmelt and rainfall samples are followed by (p), and rock transport samples are followed by (r).

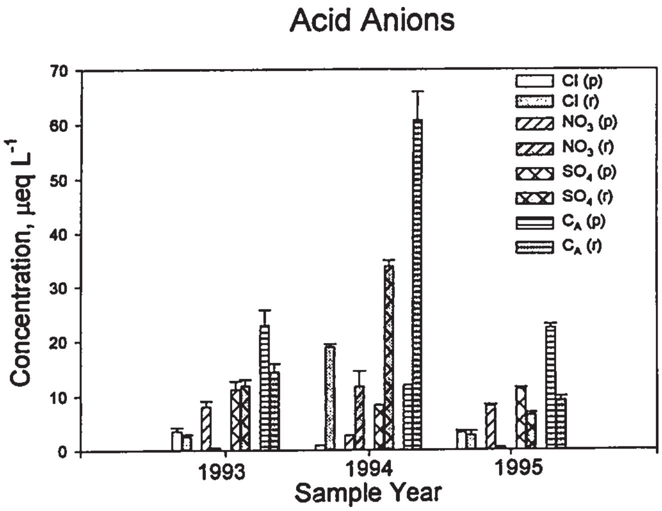

Figure 2—Concentrations of Cl, NO_3, SO_4, and sum of the acid anions (C_A) in snowmelt, rain, and runoff over rock for sampling years 1993, 1994, and 1995. Snowmelt and rainfall samples are followed by (p), and rock transport samples are followed by (r). Error bars are one standard error of the mean.

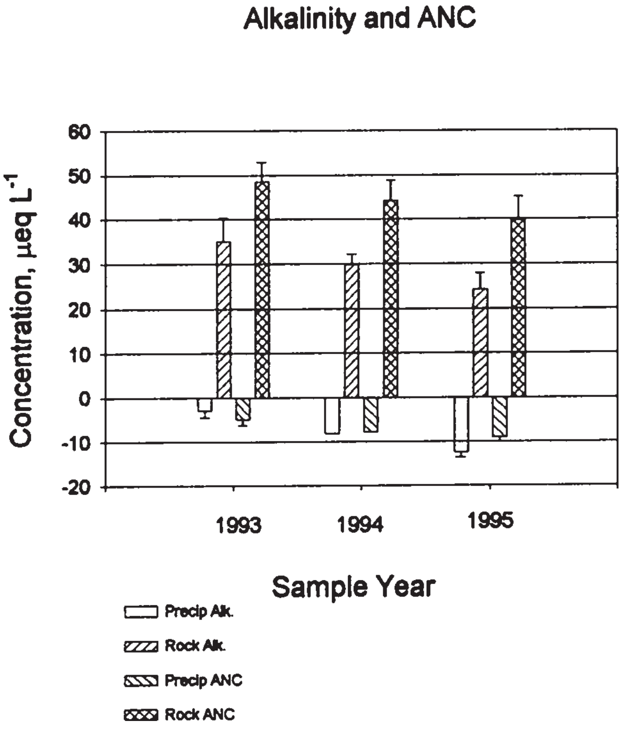

Figure 3—Alkalinity and ANC calculated as C_B-C_A for snowmelt, rain, and runoff samples. Error bars are one standard error of the mean.

for rockflow samples were consistently higher than alkalinity. Charge balance discrepancies were small for the rain and snowmelt samples, averaging –0.6 percent; however, charge balance discrepancies were consistently higher for rockflow samples, averaging +14 percent. While we assume organic ligands contribute to this difference, we did not run dissolved organic carbon analyses and have no independent check.

Effect of Rock Sampling Location

There were significant differences in concentration of Ca, Mg, C_B, alkalinity, and ANC among the five rock sampling sites. For Mg, concentration at site 1 is >2 = 3 = 4 = 5, and for alkalinity 1 >2 >3 = 4 = 5 (fig. 4). For Ca and C_B, 1 >3, and no other differences are present. These results are consistent with the expectation that greater flow path length and larger amounts of material classified as soil/vegetation would increase ANC, mainly through increased opportunity for base exchange with protons (table 3). Regressions of amount of alkalinity generated by flow over rock, calculated as the difference in precipitation and rock values for each sample period, were tested using path length, percent soil/vegetation, and the product of path length and percent soil/vegetation. There was a generally increasing trend. However, these regressions have little predictive value and are not presented. There were no significant differences in concentration of Na, K, or strong acid anions among rock sites.

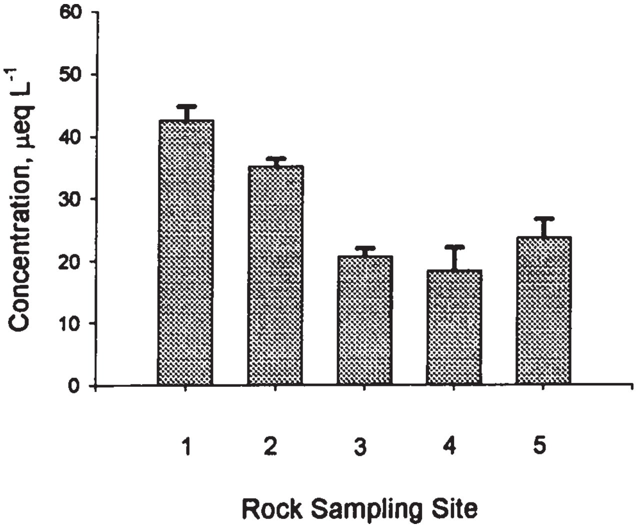

Figure 4—Average alkalinity generated at the five sample sites during rock transport for both snowmelt and rainfall events. Error bars are standard errors of the mean.

Table 3—Path length and percent of path in the categories "vegetation" or "soil" for the five rock face sampling sites.

Site	Path length	Veg/soil
	m	*Percent*
1	45	35
2	50	20
3	40	25
4	20	5
5	15	5

Discussion

The relatively small proportion of watershed surface area in soil in alpine basins suggests that other shallow ground water matrices, such as poorly sorted rock material on talus slopes and valley bottoms, are regulating stream and lake chemistry. Campbell and others (1995) reviewed a variety of studies suggesting that amorphous silicate and calcite dissolution, plus cation exchange with fast reaction times on the order of hours to days, can supply base cations that control stream chemistry. They also suggested that NO_3 concentrations are controlled by a variety of processes, including an initial concentrated pulse during melt that might be augmented by antecedent (non-snowpack) N-deposition, causing very high concentrations in stream water. There is no evidence of that in snow meltwater in the Hobbs Lake watershed.

The large and consistent decreases in $[NO_3]$ in snow meltwater (but not rain) following transport over rock (fig. 2; table 2) are interesting, both in terms of mechanism, and for their contribution to ANC reduction. Williams and others (1996) studied N transformations in snowpack and in soil in the Colorado Front Range, and reported little biological processing in snow, but rapid changes in soil. Initial meltwater in their study showed a well-defined N pulse with concentrations about 20 times bulk snowpack concentrations, declining rapidly to bulk concentrations within 20 days of melt initiation. In contrast, snowmelt and snowpack concentrations of NO_3 are similar in the Wind River Range (table 1); however, it is not clear that we sampled initial melt in 1993 or 1995. Nitrate concentrations measured at lake inlets in the vicinity of our snow sampling site do not indicate seasonal trends indicative of an "acid pulse." However, these data may not have captured initial melt either. Whether we captured an initial, elevated pulse of NO_3 or not, there clearly is some mechanism for decreasing concentration during short distance transport over rock and saprolite. At Loch Vale Watershed in the Colorado Front Range, Baron and Campbell (1997) found that rock outcrop and talus are biologically active, retain significant amounts of water, and strongly influence stream chemistry. Mass balance calculations suggest that (by difference) approximately 10 percent of atmospheric N-inputs are stored or immobilized in the bedrock compartment of Loch Vale Watershed.

Williams and others (1996) said the idea that NO_3 in surface water is from snowmelt and not soils needs to be revisited. Their results suggest that NO_3 export from subsurface environments to watercourses is controlled by the balance between gross mineralization and assimilation in soils; areas with more developed soils or extensive vegetation, or both, may have less NO_3 export. Our data suggest that in areas with low N deposition, large proportions of NO_3 are assimilated during relatively brief processing over rock outcrop and saprolite. Nitrate concentrations in Hobbs Lake sampled at the inlet, outlet, and lake surface during 1991 to 1993 averaged 1.6 $\mu eq\ L^{-1}$ (Baron 1996), and a synoptic sample in 1997 had a NO_3 concentration of 0.63 $\mu eq\ L^{-1}$ (data provided by Bridger-Teton National Forest). On the basis of these lake concentrations, one might conclude that over-rock flow during the periods we sampled may be as effective at reducing NO_3 concentrations as percolation through soil.

The other major contributor to ANC generation during over-rock transport is increased concentration of C_B. C_B concentrations averaged 14 $\mu eq\ L^{-1}$ in snowmelt and rain in this study, and increased to an average value of 58 $\mu eq\ L^{-1}$. Average lake concentration of C_B was 130 $\mu eq\ L^{-1}$ during 1991 to 1993 (Baron 1996). This might be considered an estimate of the integrated value of the net C_B provided by atmospheric and terrestrial sources, ignoring in-lake changes by biota and evapoconcentration. Lake outflow is estimated at approximately 0.8 times precipitation, and using this as an estimate of evapoconcentration provides an estimated net C_B concentration of 104 $\mu eq\ L^{-1}$ from atmospheric and terrestrial sources. Eolian contributions of fine mineral particles and salts may play an important role as a source of base cations. Dahms (1993) indicated that fine-grained volcanic minerals from the Green River Basin are found in A and B horizons of moraines on the west side of the Wind River Range. Clow and others (1997) estimated that atmospheric contributions of Ca to Loch Vale Watershed in the Colorado Front Range may range from ⅕ to ⅓ of the total Ca solute flux from weathering, exchange, and atmospheric sources combined.

Clayton and others (1991b) showed that soils in the Hobbs Lake basin have considerable capacity to neutralize H_2SO_4 inputs by base exchange during laboratory column leaching experiments, although there is no way to directly compare those lab results to the field studies of over-rock flow to estimate the relative contribution of base cations from soil and rock. However, a crude estimate can be made by comparing the lake C_B concentration, corrected for evapoconcentration, to rock-flow C_B concentration, and ascribing the difference to soil-derived C_B, when correcting for the proportion of the watershed area in rock and soil. Approximately 70 percent of Hobbs Lake basin surficial area is in rock outcrop and 30 percent is in soil (Clayton and others 1991a). This calculation indicates that per unit area, soil is approximately 3.5 times more effective at supplying base cations than flow over-rock outcrop.

Information on increases in C_B concentration from over-rock flow can be incorporated into MAGIC in more than one way; the choice depends on assumptions about the origin of the base cation supply. The source may include salts from both eolian and weathering origin (for example, $CaCO_3$, that has either blown in or dehydrated from $Ca(HCO_3)_2$ in solution as a local weathering product), exchangeable bases, and hydrolysis of primary minerals. We parameterized MAGIC so that soil compartment 2 (rock) has no depth and therefore no exchange capacity, so that all bases from the rock compartment arise from weathering. This assumes an essentially infinite source for rock-derived bases that are provided at a rate mediated by pH dependence of the weathering reaction. This is inappropriate if the main source is eolian carbonates supplied at a finite rate from a distant source. In this case, it might be more appropriate to apply an annual "treatment," which is an option in MAGIC. Forecasts under scenarios of increasing acid deposition will respond differently to these two options.

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