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Chemical fluxes and sensitivity to acidification of two high-elevation catchments in southern Wyoming

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Abstract

Hydrological and chemical fluxes were examined for East and West Glacier Lakes and their adjacent high-elevation (3200–3700 m) catchments in the Snowy Range of southern Wyoming. Both lakes are approximately 3 ha, but the East Glacier catchment (29 ha) is about half the size of West Glacier. Bedrock is primarily quartzite that has been heavily fractured and crossed with mafic intrusions.

Precipitation pH averages about 5.4–5.5, and weighted mean acid neutralizing capacities (ANC) of the discharge are about 50 μequiv.l^{-1} for East Glacier lake and 39 μequiv.l^{-1} for West Glacier, while the respective annual base cation removals are about 36 mequiv.m^{-2} and 73 mequiv.m^{-2}. Two West Glacier tributary streams average less than 10 μequiv.l^{-1} ANC, but solute concentrations during the early snow melt are more than five times those found in midsummer. It is inferred that these early high concentrations primarily are due to early elution of solutes from the snowpack rather than the displacement of high-concentration groundwater, but the cations may be substantially affected by exchange reactions.

Preliminary evaluation suggests that the mean ANC of both lakes would fall below zero if precipitation pH were to fall to 4.2–4.3. Episodic acidity during snowmelt and acidification of tributary streams would likely occur at a somewhat higher precipitation pH.

1. Introduction

To evaluate the impact of future changes in the levels of acidic deposition, it is necessary to understand the present fluxes of chemical components that contribute to both the acidity and alkalinity of a system. In this paper the hydrological and chemical fluxes for East and West Glacier lakes, two adjacent high-elevation

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catchments in the Snowy Range of southern Wyoming, and a pair of first-order high-gradient streams called Meadow and Cascade creeks are examined. The catchments are fully contained within the borders of the Glacier Lakes Ecosystem Experiments Site, GLEES (Musselman 1994). A preliminary evaluation of the sensitivity of these catchments to acidic deposition using the Henriksen model (Henriksen 1979, 1980; Wright 1983; Brakke et al., 1990) is also included. This site is similar to many of the high-elevation wilderness areas in the Rocky Mountains, and an understanding of the sensitivity of these areas to acidic deposition is essential for the formulation of rational policies for the protection of air-quality-related values.

Chemical inputs and outputs were previously evaluated for the West Glacier Lake catchment and tributaries by Rochette et al. (1988). However, because flow data and direct measurements of precipitation chemistry were largely lacking at that time, inputs were calculated from the composition of meltwater emerging from a snowfield, and output volumes were estimated. Flow measurements and chemical analyses for 1988, 1989, and 1990 are reported from both the East and West Glacier Lake catchments. The fluxes of the major cations and anions are then compared with present levels of atmospheric deposition.

2. Methods

2.1. Site description

East and West Glacier lakes are located in adjacent glacial cirque basins in the Snowy Range of southern Wyoming at latitude 41°22′30″N and longitude 106°15′30″W. East Glacier Lake lies at an elevation of 3282 m. It has a surface area of approximately 2.9 ha, a catchment area of 28.65 ha, and a volume of about 41.3×10^3 m^3. The elevation of West Glacier Lake is 3276 m. Its surface area is approximately 3.3 ha with a catchment area of 60.65 ha and a volume of about 45.2×10^3 m^3. Mean depth (volume/surface area) is 1.45 m and 1.37 m for East and West Glacier, respectively. A permanent snowfield exists at the top of the West Glacier Lake cirque. Cascade and Meadow creeks are tributary streams to West Glacier Lake, and have catchment areas of 9.08 ha and 7.68 ha, respectively.

The site is located on the southeast-facing side of a northeast–southwest-trending quartzite ridge that includes Medicine Bow Peak (3661 m). Vegetation is primarily alpine meadow, krummholtz conifer, and spruce-fir forest. While the bedrock is primarily quartzite, it is heavily fractured and crossed with weatherable mafic intrusions, primarily amphibolite, that comprise some 15–20% of the area (Rochette et al., 1988). Soils are primarily Typic Cryoborals, Dystric Cryochrepts, and Lithic Cryochrepts, although small areas of Cryumbrepts and Cryaquepts exist (Hopper and Walthall, 1994). There are also substantial areas of rock outcrops and rubbleland, which together comprise about 19% of the East Glacier and 61% of the West Glacier catchment areas. Excluding the areas classified as rock outcrops and rubbleland, soil depths in both catchments averaged 0.7–0.8 m. Effective depth is less as stones comprise about half the total soil volume (Reuss, 1994).

2.2. Flow measurements

Flows were measured using Parshall flumes installed at the outlets of East Glacier (EGO) and West Glacier (WGO) Lakes and near the mouths of Cascade Creek (CSC) and Meadow Creek (MDW). Flume details are available in Hasfurther et al. (1994). Data were recorded on strip charts and later hand-converted to daily or 6 h average readings from which flows were calculated. Visual readings recorded when samples were collected for chemical analyses were used to calculate flows when the strip charts were not operative owing to ice in the stilling wells and during the few malfunctions of the recorders. During these periods linear interpolations were used for days when no measurements were available. Diurnal fluctuations were taken into account by the averaging procedure when strip charts were used, but could not be accounted for when visual readings were used. The visual measurements were almost entirely during periods of low flow and would have little effect on total discharge.

2.3. Sampling and chemical analyses

In 1988 snow samples were collected from pits at a single location in the East Glacier catchment. Sampling details are available in Bales et al. (1990). In 1989 a total of 30 samples collected from four pits located between the East and West Glacier catchments between 24 March and 17 May were used to evaluate snow chemistry. In 1990, a total of 42 snow samples collected at three sites during the period 20 April to 18 May were used. Data from pits where either visual or analytical evidence suggested that melting had occurred were not used.

Water samples for chemical analyses were collected at the measuring flumes. Samples were collected on a daily to weekly basis in May and June, weekly in the summer, and weekly to biweekly in the autumn when flows were relatively low and changes in composition moderate. Sample numbers by years are shown in Table 1. Part of each sample was passed through a 0.45 μm millipore filter. The filtered sample was split and one portion acidified with ultrapure nitric acid for the determination of calcium, magnesium, sodium, and potassium, while the unacidified filtered sample was used for determination of chloride, sulfate, nitrate, ammonium, and dissolved organic carbon (DOC). DOC was not routinely determined on samples collected in 1990 or before 28 June 1988. Acid neutralizing capacity (ANC) and pH were determined on the unfiltered sample.

Table 1
Sample numbers by location and years

Location	1988	1989	1990
EGO	29	39	26
WGO	45	55	35
CSC	46	67	33
MDW	46	63	31

In general, the methods followed were those recommended by the United States Environmental Protection Agency (USEPA) for acid deposition studies (USEPA, 1987). ANC was determined by Gran titration using an automated Acid Rain Analysis System (ARAS), which was also used for pH. Calcium, magnesium, sodium, and potassium are currently determined using an atomic adsorption-atomic emission spectrophotometer. Earlier analyses for calcium and magnesium were done using an Inductively Coupled Plasma (ICP) emission spectrophotometer at the Colorado State University soil testing laboratory, but no differences in results owing to the change in methods could be discerned. Chloride, sulfate, and nitrate were determined by ion chromatography. Ammonium was determined using an automated flow injection analyzer. DOC analyses were performed by means of an automated infrared carbon analyzer using ultraviolet facilitated persulfate digestion.

2.4. Calculation and quality control

2.4.1. Stream samples

Data were examined for ion balance and general consistency with prior and later values. Generally ion balance, calculated as % difference,

$$\%\ \text{diff} = 100\ (\text{anions} - \text{cations})/(\text{total ions}) \quad (1)$$

(all in μequiv.l^{-1}), was required to be within -20 to $+15\%$, although occasionally larger differences were accepted for samples with low concentration. The midpoint of the acceptable range is below zero because most samples have a small anion deficit owing to the negative charge of dissociated organic acids.

For minor constituents such as Cl or NH_4, missing values were interpolated from the previous and next later values. Generally, if ion balance was unsatisfactory or a major constituent such as Ca, SO_4, or ANC was missing, the record was deleted. In a very few cases where deleting the record would create a gap of 2 or more weeks in the data, SO_4 was interpolated or an ANC value estimated from the regression of ANC on $C_B - C_A$ (i.e. sum of bases minus sum of acids).

Fluxes were calculated on a daily basis. If an interval where no samples were taken contained an even number of days, the first prior analytical values were projected forward for half the period and the first later values projected back for the remainder. If the period contained an odd number of days, the earlier value was used for the extra day. Volume-weighted means were calculated for each year. Three year means are simple means of the three annual volume-weighted means, a procedure that assumes that the fundamental replication is in years. Standard errors reported are for the 3 year means.

2.4.2. Precipitation samples

Quality control procedures for the snow pit samples were similar to those followed for the stream data. Snow pit chemistry for 1988 has been previously summarized (Bales et al., 1990, table 2), and means are taken from their report. Concentrations reported for the 1990 data are volume weighted. Appropriate water equivalent values

were not available for the 1989 data, so simple means of all samples meeting quality control criteria were used.

The NADP data (National Atmospheric Deposition Program, 1991) are from the Snowy Range site, which is located within the West Glacier catchment. Monthly deposition was calculated from the reported monthly concentrations and the reported precipitation. These deposition values were then summed and divided by the precipitation amounts for the period of interest to obtain volume weighted mean concentrations.

Table 2
Hydrological summary for East and West Glacier catchments

Catchment Year	Discharge ($m^3 \times 10^{-3}$)	Input			Stream output (mm)	Difference (mm)
		Snow (mm)	Rain (mm)	Total (mm)		
East Glacier						
1988	211	1090	115	1205	737	468
1989	165	748	190	938	576	362
1990	199	776	262	1038	695	343
Mean	191	871	189	1060	670	391
SE	14	110	42	78	48	39
West Glacier						
1988[a]	1002	2230	115	2345	1653	692
1989	866	1648	190	1838	1428	410
1990	1025	1527	262	1789	1691	98
Mean	964	1802	189	1991	1591	400
SE	50	217	42	178	82	194
Cascade Creek						
1988	181				199	
1989	173				191	
1990	211				232	
Mean	188				207	
SE	11				13	
Meadow Creek						
1988	135				176	
1989	116				151	
1990	114				148	
Mean	121				158	
SE	6.8				8.8	
Areas (ha)						
EGL — 28.65	CSC — 9.08					
WGL — 60.65	MDW — 7.68					

[a] See text for adjustments to 1988 WGO.

3. Results

3.1. Hydrology

The character of the hydrographs is illustrated using the 1989 data (Fig. 1), the driest of the 3 years. East Glacier outlet began flowing about 10 May (Julian day 130),

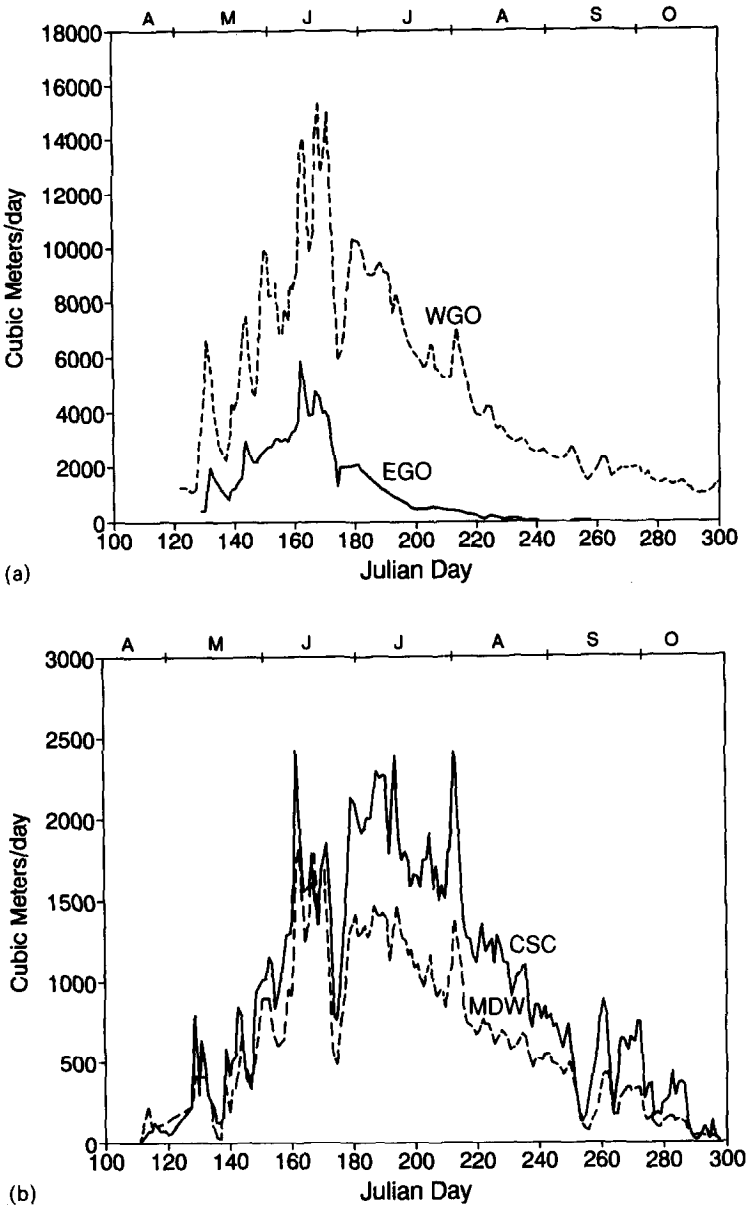

Fig. 1. Hydrographs for East and West Glacier lakes (a), and Cascade and Meadow Creeks (b) in 1989.

peaked at about 6×10^3 m^3 day^{-1} in early June, dropping to zero some time in August. In the other two years peak flows reached nearly 9×10^3 m^3 day^{-1}, and in 1988 the peak occurred at the end of June. In 1989 West Glacier outlet flow peaked in mid June at about 14×10^3 m^3 day^{-1}. In 1990 West Glacier flow reached 20×10^3 m^3 day^{-1}. Flows generally dropped below 10×10^3 m^3 day^{-1} by mid July and slowly declined throughout the late summer and early fall. Peak flows were maintained somewhat longer at West Glacier outlet than at East Glacier. Peak Cascade flows are in the range of 2.3×10^3 to 3.5×10^3 m^3 day^{-1} and occurred in late June and early July. The peak was relatively broad and the decline to near zero by mid October was nearly linear, reflecting the late melting snows at high altitude and the permanent snowfield in the catchment. Meadow Creek peak flows were in the range of 1.8×10^3 to 2.5×10^3 m^3 day^{-1}, with a general pattern similar to that of Cascade Creek.

A hydrological summary is shown in Table 2. Owing to the extreme spatial variability in snowfall amounts and the complex nature of the terrain, direct measurements of snow accumulation and water content were impractical. Therefore, snow inputs were calculated by an application of the snowmelt input term in the model of Martinec and Rango (1986). This procedure involves combining degree days with measurements of snow cover recession using aerial photography. In two separate calibration tests at the GLEES (Sommerfeld et al., 1991; R.A. Sommerfeld, unpublished data) this procedure yielded values within the 5% accuracy of a snow core probe survey on a 50 m grid.

Summer inputs are from the NADP precipitation measurements during June through September of each year. The mean West Glacier discharge for 1988 as shown in Table 2 includes an adjustment for leakage around the flume as estimated by Hasfurther et al. (1994) and for a small amount of flow that apparently occurred prior to the initial measurements. Subsequent calculations of chemical fluxes based on 1988 WGO discharge were made using the adjusted value. The leakage was repaired before the 1989 season.

Total annual discharges are reasonably consistent from year to year. Standard errors of the 3 year means range from 5 to 7% of the means for the four catchments or subcatchments. The mean annual discharge of East Glacier (192×10^3 m^3) and the volume of 41.3×10^3 m^3 result in a theoretical annual lake residence (lake volume/discharge) time of 0.215 year. The West Glacier theoretical annual residence time is only 0.046 years owing to the much higher discharge (965×10^3 m^3) and a similar lake volume. The actual residence time will vary seasonally with discharge and stratification. The large difference in discharge results from differences in area and precipitation. The area of the West Glacier catchment is more than twice that of East Glacier, and the mean precipitation inputs to the West Glacier catchment total 199 cm as compared with 106 cm at East Glacier. Winter wind velocities average 40 km h^{-1}, and the precipitation difference seems to derive largely from imports of snow owing to wind redistribution brought about by orographic effects. Apparently, imports are largely trapped in the West Glacier catchment.

The Cascade and Meadow Creek tributaries occupy 28% of the West Glacier catchment and supply 310×10^3 m^3, or approximately one-third, of the total West

Glacier Lake discharge. Flows of two other tributaries were not measured owing to the dispersed nature of the channels.

The mean difference between total precipitation input and stream output (Table 2) is 391 mm and 400 mm for East Glacier and West Glacier, respectively. This residual would include evapotranspiration (*ET*) lake evaporation, and any underground flow that would bypass the measuring flumes. Using the Blaney–Criddle method, Hasfurther et al. (1994) estimated 1988 *ET* at West Glacier at 599 mm, a value which may be somewhat high. It compares reasonably well with our 1988 residual of 692 mm but is about 200 mm in excess of the 3 year mean. There are often large uncertainties associated with water balance studies in catchments of this type (Winter, 1981), and these are reflected in the differences among years in our residuals at West Glacier. However, it is unlikely that large unmeasured underground flows are present. Such losses would tend to cause residuals in excess of *ET*, when in fact, the mean of our residuals is below estimated *ET*.

3.2. Precipitation chemistry

The two estimates of precipitation chemistry are shown in Table 3. Data from both NADP and snow pits are subject to severe limitations. Collection efficiency is very low in the NADP collectors during the winter months, often less than 10%. These low

Table 3
Summary of estimates of chemical composition (μequiv. l^{-1}) of precipitation inputs at GLEES, and at two high-altitude sites located in the Colorado Front Range

Ion	Snow-pit (3 years)[a]		NADP (4 years)		Loch Vale[b]	Green lakes[c]
	Mean	SE	Mean	SE	(Mean)	(Geom. mean)
Cations						
H^+	4.0		4.4	0.91	7.0	9.3
Ca	9.4	0.04	9.3	0.21	8.5	9.3
Mg	2.2	0.17	2.2	0.08	2.3	0.7
Na	2.4	0.06	8.0	1.11	3.3	1.8
K	1.7	0.42	0.4	0.05	0.5	2.6
NH_4	4.0	0.36	6.4	1.51	6.0	
Anions						
Cl	2.6	0.18	3.7	0.18	2.4	4.3
SO_4	9.7	1.47	15.0	1.20	13.5	13.0
NO_3	10.2	0.63	10.2	1.12	10.9	5.5
$C_B - C_A$	−2.7		−2.7		−6.2	−8.4
pH	5.4		5.4		5.2	5.0

[a] Snow-pit data include summer values (June–Sept.) from NADP.
[b] Baron et al. (1991).
[c] Caine and Thurman (1990).
Snow-pit data for 1988 taken from summary of Bales et al. (1990).

efficiencies seem to be related to snow blowing out of the collection buckets and/or to failure of the automatic opening device during snow events. Use of the NADP data then requires the implicit assumption that the snow collected is of the same composition as the snow that was not collected. The basic problem with snow pit data is the tendency for differential elution during melting. It has long been known that solute concentrations in the early melt water are much higher than those in the bulk snowpack (e.g. Johannessen and Henriksen, 1978), and this effect has been noted at the GLEES by several authors (Rochette et al., 1988; Bales et al., 1990; Finley, 1992). Thus, a very small amount of melting may alter the chemical composition. If early measurements are used in an attempt to minimize this problem, significant snow events may not be included. As summer precipitation is not included in the snow data, NADP values were used for June through September for both amount and chemical composition.

Table 3 shows reasonable agreement between snow pit and NADP results for Ca, Mg, Cl, NH_4, and NO_3. The snow pit mean annual concentration for K (1.7 μequiv. l^{-1}) is higher than the NADP mean (0.4 μequiv. l^{-1}) but in either case the precipitation input is very low and this difference is probably not very important. The NADP mean for Na (8.0 μequiv. l^{-1}) is substantially higher than the snow pit mean of 2.4. Sulfate is a key parameter and the difference between the annual values as determined from the NADP data (15.0 μequiv. l^{-1}) and that obtained from snow pits supplemented with summer NADP data (9.7 μequiv. l^{-1}) is somewhat disturbing. Table 3 includes values reported from two high-elevation sites in the Colorado Front Range, i.e. Loch Vale (Baron et al., 1991) and Green Lakes (Caine and Thurman, 1990). The Loch Vale data are from an NADP site in the Loch Vale catchment, and the Green Lakes samples were from open buckets collected on a monthly basis. For the most part the precipitation at these sites is quite similar to that found at GLEES.

The Na values (3.3 μequiv. l^{-1} at Loch Vale and 1.8 μequiv. l^{-1} at Green Lakes), are similar to the GLEES snow pit values but much lower than the GLEES NADP values. Sulfate values at Loch Vale and Green Lakes are intermediate between the GLEES snow pit and NADP values.

3.3. Surface water chemistry

A summary of the stream chemistry, averaged over years, is shown in Table 4. East Glacier Lake outlet clearly has the highest ANC, averaging about 50 μequiv. l^{-1} as compared with 39 μequiv. l^{-1} for West Glacier. These ANC values are quite low for the region. Landers et al. (1987) estimate that only 6.9% of lakes in the Central Rockies have an ANC $\leqslant 50$ μequiv. l^{-1}. However, the East and West Glacier lakes values are quite comparable to the median ANC of 42.6 μequiv. l^{-1} reported by Baron (1991a) for Loch Vale, a 5 ha lake located in the Colorado Front Range at an elevation of 3050 m in a catchment dominated by biotite gneiss. ANC values (HCO_3) reported by Caine and Thurman (1990) for the Green Lakes area, a high-elevation catchment in the Colorado Front Range where the bedrock is dominated by granites and quartz monzonites, range from 34 to 133 μequiv. l^{-1} at the various sampling points.

Table 4
Mean chemical composition for East Glacier, West Glacier, Cascade Creek and Meadow Creek

	No. years	East Glacier (μequiv. l^{-1})		West Glacier (μequiv. l^{-1})		Cascade (μequiv. l^{-1})		Meadow (μequiv. l^{-1})	
		Mean	SE	Mean	SE	Mean	SE	Mean	SE
Ions									
H^+ (Calc)	3	0.11	0.005	0.14	0.063	0.83	0.091	0.58	0.026
Ca	3	46.41	0.96	37.74	1.69	21.38	1.20	21.90	0.66
Mg	3	17.55	0.33	15.05	0.29	7.56	0.31	7.78	0.13
Na	3	10.73	0.26	8.91	0.78	5.76	0.85	6.20	0.83
K	3	2.97	0.27	3.52	0.71	1.92	0.65	2.43	0.62
NH_4	3	0.34	0.07	0.55	0.06	0.50	0.05	0.33	0.09
Sum(+)	3	78.11	1.33	65.91	3.48	37.94	2.70	39.23	1.96
Cl	3	2.85	0.77	3.10	0.25	2.60	0.33	2.66	0.35
SO_4	3	16.80	0.42	17.22	0.43	13.75	0.41	13.05	0.21
NO_3	3	0.61	0.12	4.93	1.05	11.11	1.09	9.60	0.60
[ANC]	3	50.39	2.26	38.62	2.35	5.89	0.78	8.72	0.43
Sum(−)	3	70.76	1.47	64.01	2.99	34.17	1.83	34.61	0.73
Anion deficit	3	7.35	2.31	1.90	2.66	3.77	1.42	4.61	1.48
		(mg l^{-1})	(mg l^{-1})	(mg l^{-1})	(mg l^{-1})	(mg l^{-1})	(mg l^{-1})	(mg l^{-1})	(mg l^{-1})
Other									
SiO_2	2	1.48	[a]	1.16		0.96		0.84	
DOC	2	2.24		1.26		0.90		1.54	
Charge (μequiv. mg^{-1} DOC)		3.29		1.51		4.20		3.00	

[a] SE not reported if mean contains less than three values.

The volume-weighted mean ANC of the two West Glacier tributaries is less than 10 μequiv. l^{-1}. This low ANC can be attributed to the quartzite bedrock, limited contact of these streams with the mafic dikes, and the influence of the permanent snow field. The relatively large increase of ANC at West Glacier outlet over that found in the tributary streams likely is due to the contribution of groundwater and a greater concentration of mafic dikes in the area of the two ungaged streams (Rochette et al., 1988; Finley, 1992). The ANC for samples collected from one of these (Boulder Creek) averaged 42 μequiv. l^{-1} over the 3 year period.

There is an enrichment of bases and ANC in the tributary streams and the lake outlets over that found in the precipitation input as determined from snow pits and summer NADP values. The equations for the regression of the change in base concentration on change in ANC are

$$\Delta[\mathrm{Ca}] = 6.51 + 0.557\Delta[\mathrm{ANC}] \qquad (r^2 = 0.994)$$

$$\Delta[\mathrm{Mg}] = 3.16 + 0.231\Delta[\mathrm{ANC}] \qquad (r^2 = 0.998)$$

$$\Delta[\mathrm{Na}] = 2.03 + 0.099\Delta[\mathrm{ANC}] \qquad (r^2 = 0.925)$$

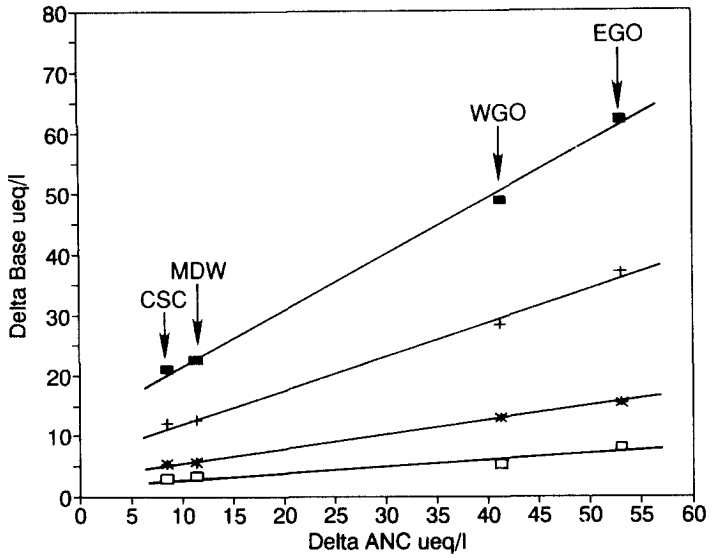

Fig. 2. Regression of change in sum of bases, Ca, Mg, and Na concentrations on change in ANC. Changes were calculated using mean snow-pit chemistry as input.

$$\Delta[\mathrm{K}] = 0.83 + 0.025\Delta[\mathrm{ANC}] \qquad (r^2 = 0.669)$$

$$\Delta[\text{sum bases}] = 12.53 + 0.912\Delta[\mathrm{ANC}] \qquad (r^2 = 0.997)$$

While only four points are available for each line, the fit for Ca, Mg, and sum of bases is very good ($P < 0.01$). This need not surprise us because the enrichment of bases from sources in the catchment is the major process contributing to the generation of ANC. The relationship for Na is not as good ($P < 0.05$) but still indicates a contribution of Na from the catchment to lake ANC. The contribution of catchment K to lake ANC cannot be shown to be significant.

The slopes provide an estimate of the relative contribution of each component to the total ANC generation. While the ANC in stream or lake waters increased from about 9 to 55 μequiv. l^{-1} over the ANC of the snow, some 56% of this increase is due to Ca enrichment, 23% to Mg, and 10% to Na. The relationships are shown graphically in Fig. 2. The ratio of the slopes for Ca and Mg also shows that a consistent 2.4 times as much Ca as Mg is weathered from the catchments. This may be compared with a Ca/Mg ratio of 3.9 in the snow pack, 2.8 in the tributary streams and 2.5 at the West Glacier outlet, indicating that the ratio approaches the weathering ratio as the sampling points move further downstream where the impact of weathering of the mafic dikes increases.

The intercept for the sum of the bases indicates that an initial enrichment of about 12.5 μequiv. l^{-1} (SE = 1.2 μequiv. l^{-1}) of base cations occurs without a concurrent increase in ANC; i.e. processes are occurring that result in lower ANC than would be expected strictly on the basis of the increase in bases. The loss of nearly 4 μequiv. l^{-1} NH_4 between snowpack and the sampling points on Cascade and Meadow creeks is

undoubtedly a factor. This could be the result of cation exchange of Ca and Mg for NH_4, or from loss of ANC owing to release of H^+ owing to either the uptake of NH_4 or oxidation of NH_4 to NO_3. Another likely factor is the formation of organic anions, estimated by charge deficit at approximately 4–5 μequiv. l^{-1} in these creeks. These total slightly less than the observed difference, a discrepancy that may be due to random error, analytical error, or unidentified processes.

The slope of the sum of bases line (0.912, SE = 0.036) is less than 1.0, implying a source of ANC which does not result in increased base cation concentration. This is most likely due to biological removal of NO_3. The NO_3 levels in Meadow Creek and Cascade Creek are 11.1 μequiv. l^{-1} and 9.6 μequiv. l^{-1}, respectively, about the same as those found in the precipitation. The average lake outlet concentrations are 4.9 μequiv. l^{-1} and 0.6 μequiv. l^{-1} for WGO and EGO, respectively. This implies virtually complete immobilization of NO_3 in East Glacier and partial immobilization in West Glacier Lake. The greater immobilization of NO_3 in East Glacier simply may be due to the longer residence time but it may also reflect a higher level of biological uptake of NO_3 in East Glacier, as suggested by the higher observed DOC, i.e. 2.2 mg l^{-1} at EGO and 1.3 mg l^{-1} at WGO.

The observed NO_3 levels are below the median value of 16.4 μequiv. l^{-1} reported by Baron (1991a) for Loch Vale. They are also generally lower than those found by Caine and Thurman (1990) in the Green Lakes area, where means ranged from 3.5 to 22.7 μequiv. l^{-1}. The lowest values in the Green Lakes area were observed at the outlet of the largest lake, which was located low in the watershed, with higher NO_3 values at the higher sampling points. This trend is consistent with that observed at GLEES but was much less pronounced at Loch Vale.

The observed mean SO_4 concentrations of ca. 17 μequiv. l^{-1} (Table 4) are very near the same at both EGO and WGO. Again, these are lower than most lakes in the Central Rockies where Landers et al. (1987) estimates that 20% of the lakes are below 19.1 μequiv. l^{-1}. However, judging from the frequency distributions of Turk (1988) and Turk and Spahr (1991), the SO_4 concentrations are slightly below the median for lakes in the Colorado Front Range to the south, and slightly above the median for lakes in the Mt. Zirkel Wilderness to the southwest. The mean SO_4 concentrations in the Cascade and Meadow creeks was 13.8 μequiv. l^{-1} and 13.1 μequiv. l^{-1}, respectively. As with NO_3, the SO_4 concentrations at GLEES are substantially below the median Loch Vale value of 36.0 μequiv. l^{-1} (Baron, 1991a), and below most of the values reported for Green Lakes. However, concentrations reported for various sampling points within the catchment at the latter site were highly variable, with means ranging from 9.2 to 85.6 μequiv. l^{-1}.

Seasonal changes in sum of bases (C_B), sum of acids (C_A), ANC, and pH are illustrated in Figs. 3 and 4, using the 1989 data. At East Glacier Lake, initial concentrations tend to be high but somewhat variable. Concentrations then decline until about 1 July, followed by an increase later in the season. The high early ANC values apparently are due to in-lake generation of ANC over winter, perhaps accompanied by an influx of high ANC groundwater. As melting proceeds, concentrations drop owing to dilution. Later in the season concentrations again rise as the proportion of surface inflow decreases and the groundwater influence increases. During this period

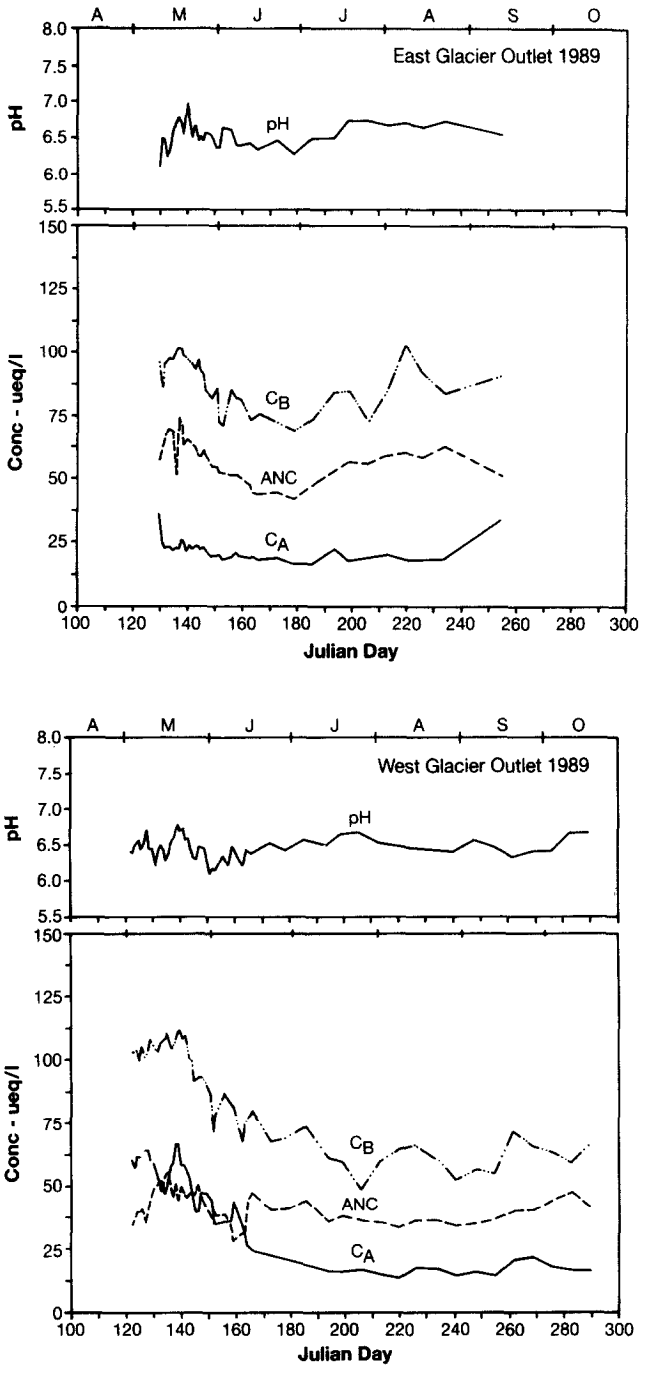

Fig. 3. Sum of bases (C_B), sum of acids (C_A), ANC, and pH by Julian Day for East and West Glacier outlets in 1989.

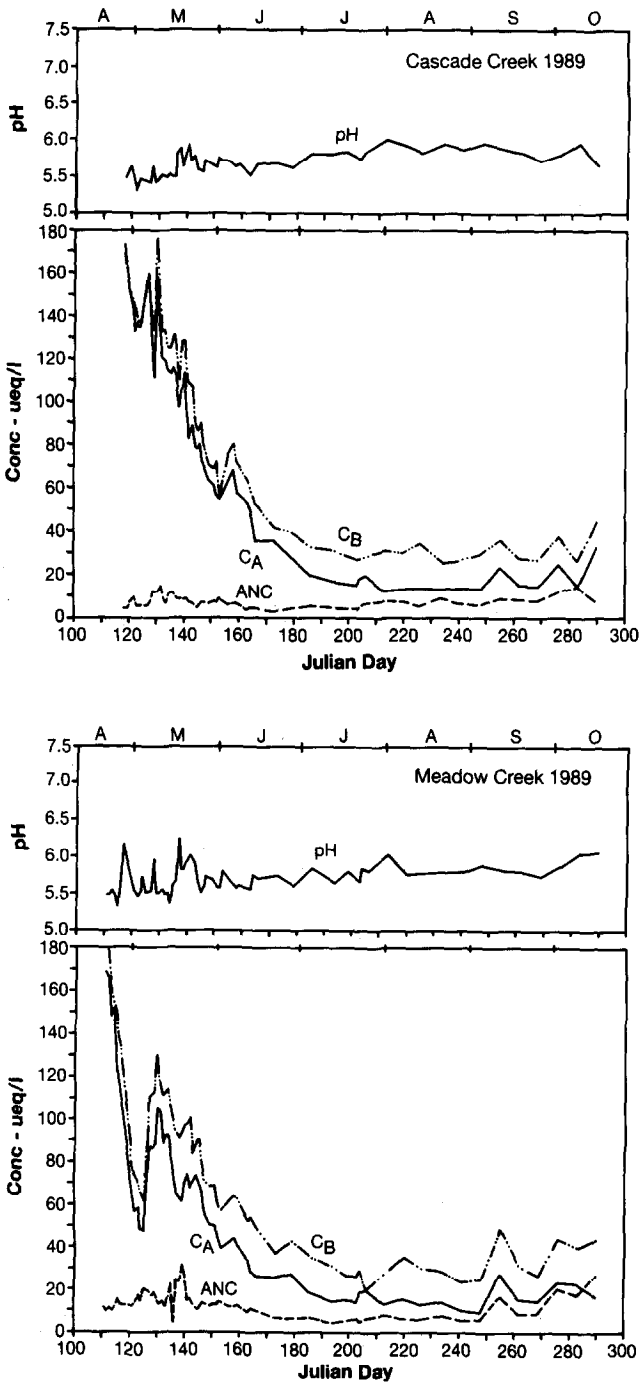

Fig. 4. Sum of bases (C_B), sum of acids (C_A), ANC, and pH by Julian Day for Cascade and Meadow creeks in 1989.

the increase in bases (Ca, Mg, Na, K, and NH_4) is greater than the increase in strong acid-forming anions (SO_4, NO_3, and Cl), so ANC and pH increase.

West Glacier outlet (Fig. 3) also exhibited high but variable early season solute concentrations with a marked decrease in midseason. The high early values for C_B and C_A again likely result from a combination of influx from the ground water during the winter and high concentrations in the early meltwater. High early ANC values are

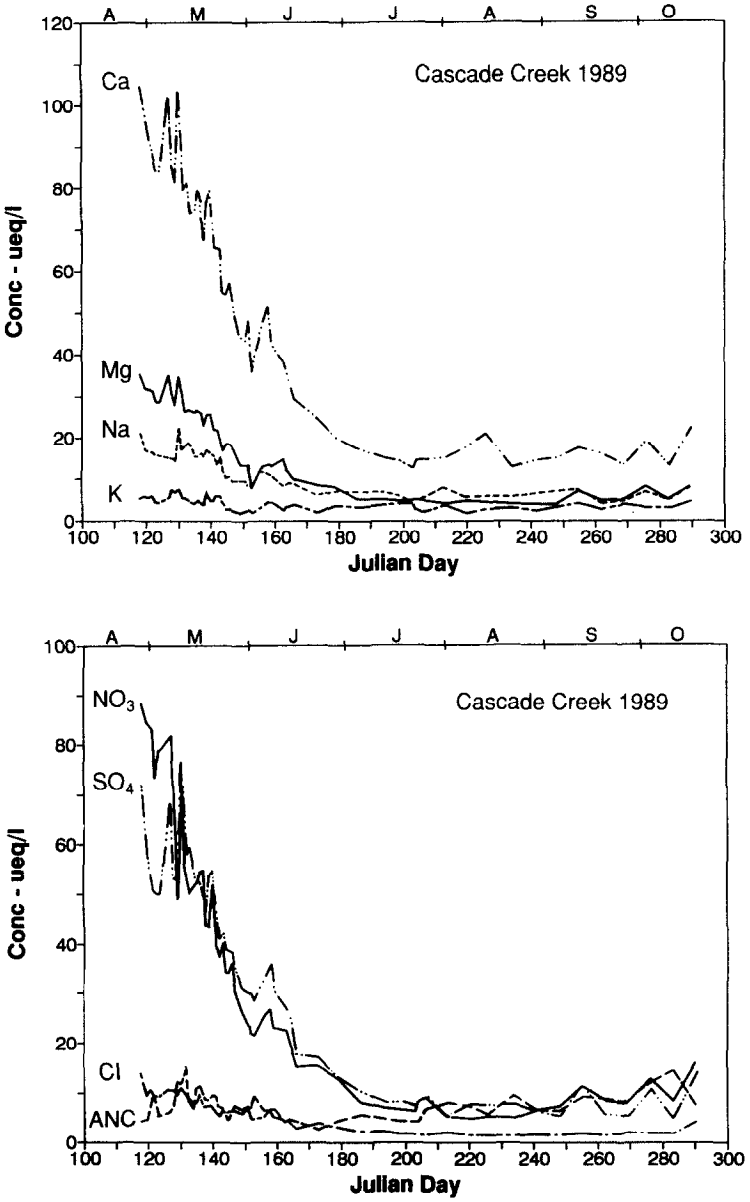

Fig. 5. Concentrations of individual ions by Julian Day for Cascade Creek in 1989.

due at least in part to in-lake processes over the winter (Vertucci, 1991). A drop in ANC and pH was observed in late May and early June. This drop was more pronounced in both 1988 and 1990 (not shown) than in 1989. It can be attributed to a tendency for the bases (C_B) to drop more rapidly than do the acids (C_A) resulting in a marked decrease in pH and ANC. The 1988 drop in West Glacier lake has been interpreted as supporting a small pulse of episodic acidification owing to chemical balance in addition to the drop in pH that can be attributed to dilution (Vertucci, 1988). At West Glacier low concentration streamflow from the high snowfields supplements the higher concentration groundwater inflow during the late season, so the late season rise in ANC is less noticeable than in East Glacier. However, an increase in ANC and pH occurs at about day 170 in all 3 years, apparently as a result of an influx of soil water and groundwater.

High solute concentrations during early snowmelt are very evident in the tributary streams (Figs. 4 and 5), although early values from Meadow Creek were somewhat erratic in 1989. The question arises as to whether these high early concentrations reflect the differential melting of the snowpack or whether 'piston flow' is displacing high concentration water from the soil and rock interstices. There is a small increase in bases and ANC when compared to precipitation input, indicating that some bases are furnished by weathering and/or soil exchange. Time traces for individual ions in Cascade Creek in 1989 as shown in Fig. 5 are typical. Early concentrations are very high, particularly for Ca, NO_3 and SO_4. High early concentrations of Ca and Mg could conceivably result from weathering and displacement by early meltwater, but if this were the case correspondingly high levels of ANC would be expected. However, the major anions are SO_4 and NO_3, which are much more likely to arise from atmospheric sources. The 3 year weighted means for SO_4 and NO_3 in the snow

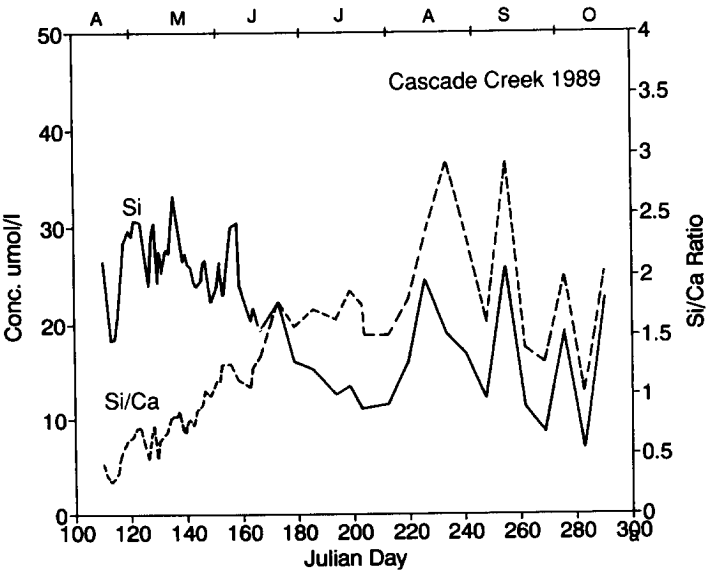

Fig. 6. Concentration of Si (μmol l^{-1}) and Si/Ca ratio in Cascade Creek in 1989.

pack are 9.7 μequiv. l^{-1} and 10.2 μequiv. l^{-1}, respectively. The high early levels of SO_4 and NO_3 that track closely at very nearly equal concentrations are strong evidence for early elution of solutes from the snowpack. This is supported by the early elution observed in snow lysimeters by Bales et al. (1990). Further evidence is available from the Si concentrations and Si/Ca ratios shown in Fig. 6. Silica values do tend to be high in spring, suggesting that at least some of the water may come from the soil and rock pores. However, this seasonal pattern is not nearly as marked for Si as it is for Ca, so molar Si/Ca ratios in the spring are between 0.5 and 1.0, increasing to about 2.5 over the course of the season. As the Si/Ca ratios are lowest during the period of highest

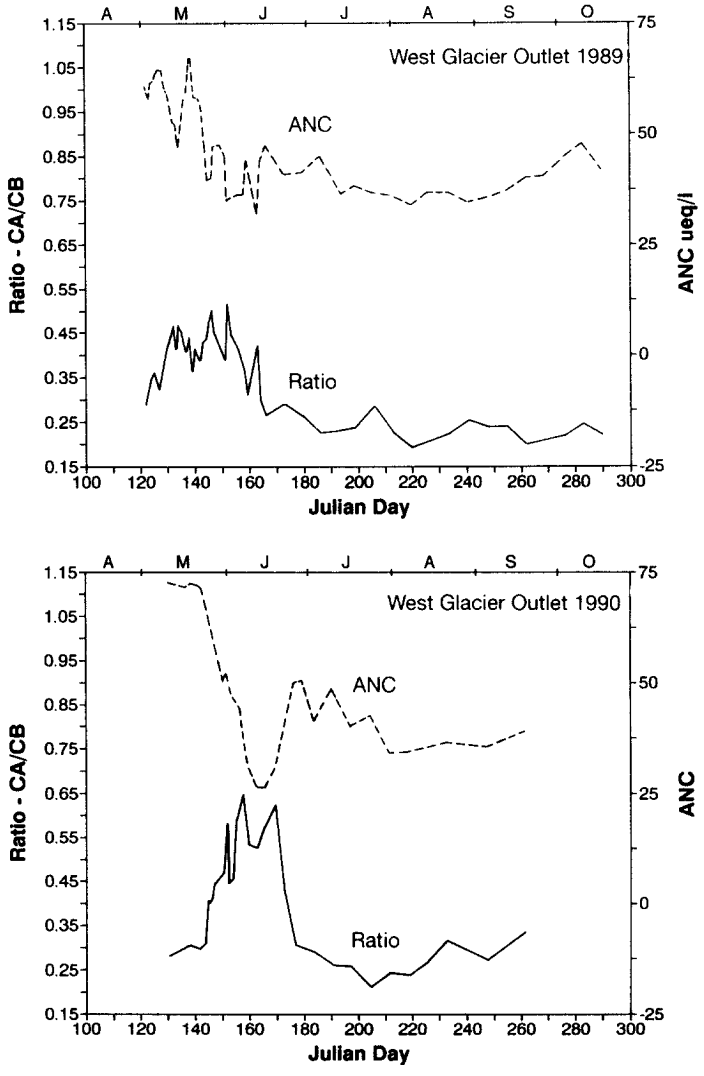

Fig. 7. Time series of ANC and the ratio of (SO_4 + NO_3) to (Ca + Mg + Na + K) for West Glacier Lake outlet in 1989 and 1990.

concentration of other solutes, much of the high Ca concentration during the early season is likely the result, either directly or indirectly, of differential release of solutes from the snowpack during melting.

We have also examined time series of the ratios of the strong acid-forming anions ($SO_4 + NO_3$) to base cations (Ca + Mg + Na + K). The mean ratio in the snowpack is 1.27, and as base cations are picked up from the catchment ANC increases and the ratio decreases. If the high early solute concentrations are due to the direct influence of differential elution during snowmelt, the ratio should increase at that time, while if it were due to deeper water displaced by piston flow this increase would not occur.

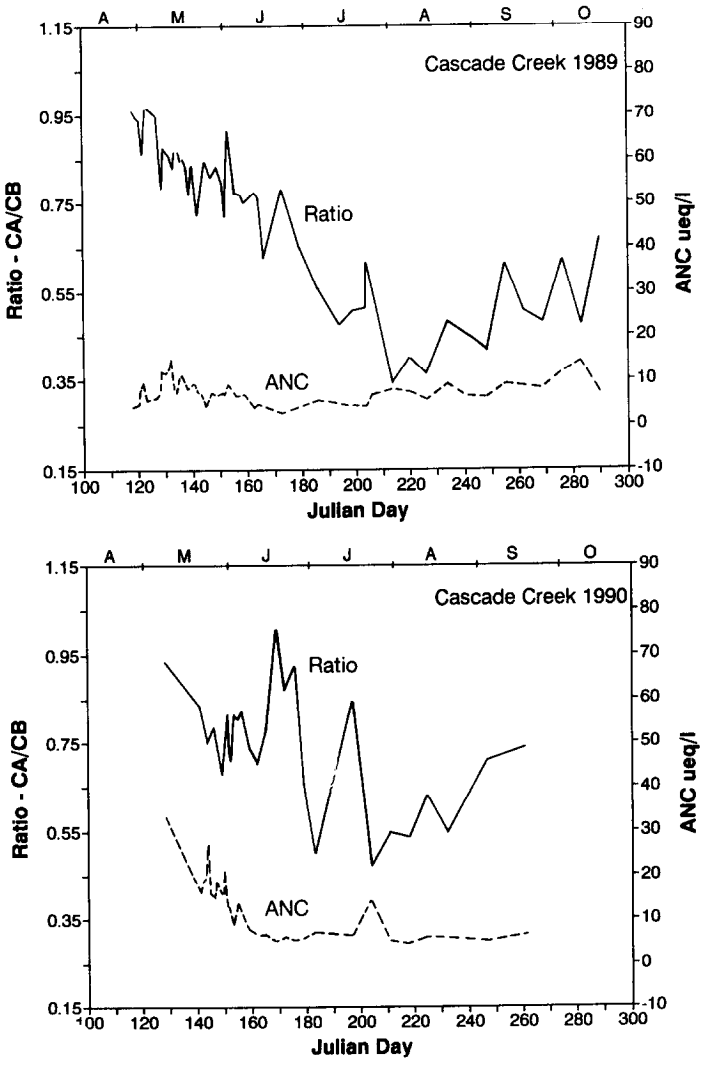

Fig. 8. Time series ANC and the ratio of ($SO_4 + NO_3$) to (Ca + Mg + Na + K) for Cascade Creek in 1989 and 1990.

A clear signal of increased ratios corresponding to a loss of ANC was detected in West Glacier Lake in all 3 years, and is illustrated by the 1989 and 1990 values shown in Fig. 7. The signal was particularly strong in 1990. Simple dilution during snowmelt would tend to reduce ANC without having much effect on this ratio, while episodic loss of ANC owing to the influence of snow chemistry would tend to increase the ratio owing to the higher ratio found in the snow. No such signal was observed in East Glacier Lake in any of the 3 years, apparently owing to greater interaction of the snowmelt with soil and groundwater during snowmelt.

The ratios in the tributary streams are much higher and ANC values much lower than those at the lake outlets (Fig. 8), reflecting a greater influence of snow chemistry on the surface water. The patterns were also much more variable, both within and among years, than those observed in West Glacier Lake. The higher within-year variability can be attributed to the lower base concentrations, as small variations in the denominator will have a disproportionate effect on the ratio, while the between-year variability likely is due to differences in melt patterns. Even so, a signal can be discerned in the 1990 values, with a peak in the ratios coinciding with an early drop in ANC. In 1989 ANC was already near its lowest point at the time of the initial sampling, and while initial ratios were very high indicating a major influence of meltwater one can not actually discern a drop in ANC coincident with a peak in the ratios. The concentration of both bases and acids in these tributary streams is high early on and drops off as melting proceeds.

While the above clearly indicates that the chemistry of the lakes and particularly the low ANC tributaries are heavily influenced by differential elution of solutes during snow melt, it should not be concluded that there has been little or no interaction with the catchment, as important chemical changes can occur as a result of only limited contact. The higher absolute Si concentrations during the early season suggest some mixing with old water in the catchment. Also, cation exchange may have taken place, and the fact that base cation concentrations in the low ANC tributaries closely track the strong acid anion concentrations would be consistent with a model in which the anions are primarily controlled by the meltwater, while the cations may be heavily influenced by exchange processes (Dahl et al., 1980; Seip, 1980). Support for the influence of exchange on cations is provided by the fact that throughout the season the Mg/Ca ratios in the streams exceed those in the snowpack.

3.4. Chemical fluxes

Summaries of the 3 year mean chemical fluxes are shown in Table 5. Yearly inputs are based on the precipitation amounts measured for that year but input concentrations are averaged over years (Table 3) prior to use in the calculations. This procedure was adopted because the variability of precipitation chemistry measurements within years is greater than variability among years. Thus, concentration differences among years are unlikely to be meaningful, and the best estimates are probably those that include as many observations as possible. Yearly outputs are based on measurements of both flow and concentration made in that year. Yearly

Table 5
Three year mean input and output summaries for East and West Glacier catchments using snow-pit input

Ion	Input (mequiv. m^{-2})	Output (mequiv. m^{-2})	Diff. (mequiv. m^{-2})	SE Diff. (mequiv. m^{-2})
East Glacier				
H^+	4.3	0.1	4.2	0.31
Ca	10.0	31.0	−21.0	1.04
Mg	2.4	11.8	−9.4	0.88
Na	2.5	7.2	−4.6	0.21
K	1.8	2.0	−0.2	0.13
NH_4	4.3	0.2	4.0	0.34
Cl	2.7	2.0	0.7	0.45
SO_4	10.3	11.2	−1.0	0.06
NO_3	10.8	0.4	10.4	0.82
ANC	−2.9	33.6	−36.5	2.36
West Glacier				
H^+	8.0	0.3	7.7	0.75
Ca	18.7	59.8	−41.1	2.29
Mg	4.4	23.9	−19.5	1.01
Na	4.7	14.0	−9.3	0.98
K	3.3	5.5	−2.2	1.10
NH_4	8.0	0.9	7.1	0.81
Cl	5.1	4.9	0.2	0.19
SO_4	19.3	27.3	−8.1	2.01
NO_3	20.2	8.0	12.3	3.27
ANC	−5.4	61.2	−66.6	3.23

Negative values indicate a net removal from the catchment.

inputs, outputs and fluxes are then averaged to obtain the 3 year means and standard errors.

While there is no completely satisfactory basis for choosing between snow pit and NADP input data, the budgets shown in Table 5 are based on the snow pit input (Williams and Melack, 1991). Also mass balances based on the NADP data indicate a net accumulation of Na, SO_4, and Cl in the catchments, while those based on snow chemistry input indicate a net removal of Na and SO_4, with inputs and outputs of Cl approximately in balance. The latter case seems more likely as some weathering of Na would be expected and some pyrite has been identified at least in the West Glacier catchment (Rochette et al., 1988; Finley, 1992). Fortunately, there is substantial agreement between the fluxes of Ca, Mg, K, and NO_3, using either set of input data.

The only dry deposition included in the input fluxes is that which accumulates in the snowpack, (snow-pit data only), so that the input may be underestimated. This limitation is not as serious here as in many systems because the primary precipitation input comes from the snowpack (Table 1). Also, based on the soil maps of Hopper and Walthall (1994), only 39% of the West Glacier catchment is vegetated, much of

which is tundra or krumholtz that is covered by snow for most of the year, reducing the surface area for collection of dry deposition.

3.4.1. Base cations

As shown in Table 5, net Na removal is estimated to be 4.6 mequiv. m^{-2} and 9.3 mequiv. m^{-2} for East Glacier and West Glacier, respectively. Total annual base cation (not including NH_4) removal from the West Glacier catchment is 72.2 mequiv. m^{-2}. This is substantially above the 44.4 mequiv. m^{-2} value estimated by Rochette et al. (1988).

Base cation removal (not including NH_4) from the East Glacier catchment is estimated as 35.2 mequiv. m^{-2}. The low flux from the East Glacier catchment is associated with lower precipitation inputs, i.e. a 2 year mean of 1060 mm at East Glacier as compared with 1991 mm at West Glacier, suggesting that base cation removal is nearly proportional to precipitation amount. This proportionality indicates that caution should be exercised in the use of cation export values to compare sensitivity of different catchments to acidic deposition (Rochette et al., 1988). On this basis, East Glacier would be more sensitive than West Glacier to an increase in deposition acidity. However, the ANC of East Glacier is higher than in West Glacier, and the analysis below suggests that the West Glacier would be somewhat more sensitive to acidification than the East Glacier. Baron (1991b) reported a net annual cation export of 51.9 mequiv. m^{-2} for the Loch Vale catchment, or about midway between that of East and West Glacier lakes. However, the molar ratio of Mg to Ca was about 3.8, compared with 2.4 for GLEES. Caine and Thurman (1990) report a rate of about 100 mequiv. m^{-2} at Green Lakes.

The NH_4 deposition in the East Glacier catchment is estimated as 4.3 mequiv. m^{-2} (0.60 kg N/ha). Deposition, estimated at 8.0 mequiv. m^{-2} (1.1 kg N/ha), is higher at West Glacier owing to the higher rainfall. Almost all of this is retained in both the East and West Glacier catchments. The low NH_4 levels in Cascade and Meadow creeks suggests that most is retained by the terrestrial or wetland components and does not actually reach the lakes.

3.4.2. Acid-forming anions

Using the snow pit input data, net annual removal of SO_4 is estimated to be 8.1 mequiv. m^{-2} or about 1.3 kg S/ha from the West Glacier catchment. However, if the NADP data were used a net retention of 2.6 mequiv. m^{-2} or 0.42 kg S/ha would be estimated. The corresponding values for East Glacier are a removal of 1.0 mequiv. m^{-2} (0.16 kg S/ha) using snow pit data or a retention of 4.7 mequiv. m^{-2} (0.75 kg S/ha). As mentioned above, some pyrite has been identified in the West Glacier catchment, which would provide a source of sulfur. On the other hand sulfur might be retained by the biota or by soil adsorption, so either net removal or accumulation is possible. The close correspondence between SO_4 and NO_3 concentrations in the Cascade (Fig. 5) and Meadow Creek (not shown) tributaries suggests that watershed sources of S are not important in these areas, but they may exist in other parts of the West Glacier catchment. While this cannot be resolved at

present, the amounts are small in each case and the effect on calculations of the effect of increased acidic deposition will be moderate at most.

The annual estimated input of NO_3 is 10.8 mequiv. m^{-2} (1.6 kg N/ha) in the East Glacier catchment and 20.2 mequiv. m^{-2} (2.8 kg N/ha) at West Glacier. Virtually no NO_3 is discharged from East Glacier, but West Glacier Lake discharges more than one-third (8.0 mequiv. m^{-2}) of the NO_3 input.

3.4.3. Acidity and ANC

The Henriksen model (Henriksen, 1979, 1980) as modified by Brakke et al. (1990) is used to estimate the change in precipitation acidity that would be required to acidify East and West Glacier lakes, and Cascade and Meadow creeks. In this case the end point is defined as an annual volume weighted mean ANC of zero. The Henriksen model uses an F factor that may be defined as the fraction of increased acidity that is neutralized by an increase in release of base cations from the catchment, either by accelerated weathering or soil exchange. Unfortunately, there is no really satisfactory method of predicting this value. However, unless ANC levels are very high, the F-values can be expected to be less than 1.0, owing to the physical-chemical processes by which increased solution concentration results in decreased pH of the soil solution, i.e. the 'salt effect' (Reuss and Johnson, 1986). Pointing out that lakes in catchments that are well supplied with exchangeable and/or weatherable bases will naturally have high ANC, Brakke et al. (1990) proposed that the F-value will be 1.0 in catchments that are sufficiently well supplied with bases so that virtually all acidic deposition will be neutralized, and that this will generally occur in the range of 200–400 μequiv. l^{-1} ANC. They then proposed a relationship by which the F-value may be estimated from lake ANC

$$F = \sin 90(BC^*/S), \tag{2}$$

where BC^* is the concentration of non-marine base cations and S is the ANC at which $F = 1.0$. In our case the marine contribution is small, so BC^* may be taken as simply the base cation concentration. Using this relationship, F has been calculated for S-values in the range of 200–400 μequiv. l^{-1}. These range from 0.302 to 0.575 for East Glacier and 0.256 to 0.494 for West Glacier Lake. Wright (1983) estimated a value of 0.4 for North American lakes in sensitive areas. Based on these F-values, acidification of East Glacier Lake would require an increased rainfall acidity of 45–75 μequiv. l^{-1} or a final precipitation pH of about 4.11–4.33. Acidification of West Glacier would require increased precipitation acidity of 41–61 μequiv. l^{-1}, or a final precipitation pH of 4.20–4.35. These values are similar to current or recent precipitation acidity in the Adirondack Mountains where the more sensitive lakes have become acidified (Linthurst et al., 1986; Asbury et al., 1989). Thus, while these systems must be considered moderately sensitive, they are less so than would be inferred from the earlier work of Rochette et al. (1988).

The above analysis considers only annual averages. In West Glacier Lake where there is a clear signal of episodic loss of ANC at the current low level of acidic input, acidic episodes would likely occur at somewhat higher pH values as acidity is released from the snowpack during the early stages of melting.

Cascade and Meadow Creeks appear to be much more sensitive than are the lakes. According to this analysis F-values are in the range of 0.15–0.3 and precipitation acidity of less than 10 μequiv. l^{-1}, corresponding to a precipitation pH of about 4.9, would be sufficient to reduce annual volume-weighted mean ANC values to zero. While it is entirely possible that these F-values are underestimates, even if the true values are a relatively high 0.75, the systems would acidify at precipitation pHs in the range of 4.50–4.65. Thus, these tributary streams appear to be very sensitive to acidic deposition.

Given current sources of precipitation acidity, the question arises as to what the effects would be if the precipitation were enriched with nitric acid rather than sulfuric acid. As most NO_3 currently entering the Cascade and Meadow creek subcatchments is discharged to West Glacier Lake, presumably most of the increase in NO_3 would also be passed on. Therefore, the most likely scenario for these tributary streams is that the effect on stream ANC would be similar for either sulfuric or nitric acid-enriched precipitation.

The effect of nitric acid on the acid–base status of East and West Glacier lakes is more difficult to predict. Biological immobilization would likely attenuate at least part of the acidifying effect of nitric acid inputs. Currently, the net immobilization in the West Glacier system is about two-thirds of the NO_3 input. The fact that a significant amount is currently being discharged may indicate that the lake productivity is limited by factors other than nitrogen or that the rapid turnover does not allow sufficient time for biological immobilization. In either case the system likely would not have the ability to adsorb substantial increases, so that decreased ANC would be expected, particularly during snowmelt. In the East Glacier catchment almost all NO_3 inputs are currently being immobilized and there is more opportunity for the terrestrial component of the ecosystem to utilize incoming N before it reaches the lake. To what extent increases in NO_2 can be adsorbed would be difficult to predict. In either catchment, significant increases in NO_3 inputs would most likely result in substantial changes in the species composition and productivity of both the aquatic and terrestrial systems.

4. Conclusions

Precipitation inputs and depth of runoff were markedly different in these two adjacent catchments, apparently primarily owing to elevation and differences in snow imports caused by orographic effects. The higher precipitation and runoff resulted in a lower mean ANC (39 μequiv. l^{-1}) at West Glacier than at East Glacier (50 μequiv. l^{-1}).

Solute concentrations in runoff were very high early on in the snowmelt, particularly in the first-order, high-gradient tributaries, reflecting differential elution of solutes during snow melt. The anions, mostly SO_4 and NO_3, were apparently derived primarily from the snowpack, but ion exchange may play a significant role in controlling cation concentrations.

A preliminary analysis suggests that these lakes would be moderately sensitive to

acidic deposition. Complete loss of ANC would be unlikely unless deposition acidity reached or exceeded levels similar to those found in areas of the eastern US where lakes' sensitive catchments have become acidic. However, episodic acidification during snowmelt, particularly in West Glacier Lake, and acidification of the low ANC tributaries may well occur at lower levels of acidic inputs. West Glacier Lake had higher base cation export (73 mequiv. m^{-3}) than did East Glacier (36 mequiv. m^{-3}), even though ANC is lower at West Glacier. This indicates that base cation removal rates may not be an appropriate indicator of sensitivity to acidic deposition.

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