Watershed Factors Affecting Stream Acidification in the White Mountains of New Hampshire, USA

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ABSTRACT / The streams tributary to acidic Cone Pond, pH 4.5–4.8, and circumneutral Black Pond, pH 5.3–6.4, in the White Mountains of New Hampshire, USA, were monitored for a year. The watersheds of these two ponds were characterized in terms of geology and stream hydrology. Chemical gradients and patterns in rock weathering and groundwater discharge explain many of the differences in mineral content and acidity of the streams. The rocks of Black watershed produced an average of ten times the equivalent of basic cations as rocks from Cone watershed. This is on the same order as the difference in acidity of the two streams. Downstream changes in stream chemistry follow differing patterns, but reflect the same principle of residence time and water path length controlling chemical evolution of streamwater. Watershed and aquatic managers may use these parameters in an inexpensive and simple assessment of the susceptibility of individual streams and ponds to acidification. A method is recommended to determine quickly the potential influence of bedrock type to aquatic chemistry.

The White Mountains of New Hampshire, as well as much of New England in the United States, have been identified as having aquatic systems particularly susceptible to acid deposition (Omernik and Powers 1982). However, in a study of six small ponds located in close proximity within the White Mountains, Buso and others (1984) found susceptibility to be highly variable. Our study examined watersheds of two of these six ponds, Black and Cone, which are at opposite extremes in terms of acidity. Objectives were: (a) to determine whether there were easily identified features that aquatic managers could use as indexes of susceptibility of aquatic systems to acid precipitation, and (b) to determine whether the chemical gradient in streams at these two watersheds followed an acid rain neutralization model proposed by Johnson and others (1981).

The model of Johnson and others (1981) was based on studies of stream chemistry along an elevational gradient at Falls Brook within the Hubbard Brook Experimental Forest in New Hampshire. The study illustrated the relationships between meteoric, pedologic, and geologic origins of stream water solutes and the effect of residence time in the soil-channel system on the proportions of each. At Falls Brook, acid components in upper stream reaches were replaced by chemical weathering products in lower reaches, particularly Ca^{2+} and Na^{+}, as path length of water in the soil-channel system increased. In our study, we hypothesized that flow paths in the watershed of Black Pond might be quite different from those in the Cone Pond watershed, and could thus affect stream and pond chemistry.

KEY WORDS: Acid precipitation; Acidification; New Hampshire; Ponds; Rock weathering; Stream chemistry; Streams; White Mountains

Johnson and others (1981) discussed neutralization of acids by chemical weathering. The opposite may be true in some watersheds, however, as revealed by Parnell (1983), who documented production of SO_4^{2-} by weathering of metasedimentary rocks containing small amounts of sulfides. According to Parnell's model, four equivalents of acidity are generated by dissolution of 1 mol of FeS_2. We initially suspected that weathering of sulfides might account for high SO_4^{2-} and acidity levels observed at Cone Pond.

Study Area and Methods

Site Description

Physical characteristics of the study areas are listed in Table 1. Each pond has one major inlet with 2–3 branches above a wetland area. At Black, the inlet branches above a spruce flat which has been flooded by beavers. At Cone, the inlet branches above a peatland dominated by sphagnum and yellow birch. Both

Table 1. Study area description.

	Black Pond	Cone Pond
Location	N 44°06', W 71°35' Lincoln, NH	N 43°54', W 71°36' Thornton, NH
Pond area	1.7 ha	3.1 ha
Volume	92,000 m^3	101,000 m^3
Volume weighted pH	5.3–6.4	4.5–4.8
Volume weighted bicarbonate alkalinity	60–70 μeq $liter^{-1}$	0 μeq $liter^{-1}$
Watershed area	31 ha	63 ha
Watershed aspect	east–southeast	south–southeast
Watershed mean slope	23%	24%
Watershed elevation	481–634 m	481–649 m
Watershed cover types		
Northern hardwoods	90%	20%
Conifer	10%	60%
Bedrock outcrop	0%	20%
Watershed land use	Cutover circa 1900 Uninhabited Light camping/hiking	Burned circa 1820 Uninhabited Light camping/hiking

branches of the Black Pond inlet arise from springs. The inlet to Cone Pond flows through several rocky gorges, and over short waterfalls.

The Black Pond watershed is completely mantled by glacial till. In contrast, Cone Pond watershed has about 20% exposed bedrock and the remaining area covered with glacial till. The soils of both watersheds are characterized by sandy spodosols.

Stream Sampling

Stream samples were collected at seven points along each inlet stream. In both watersheds, two branches of the inlet were sampled. Sampling was conducted from June 1983 through May 1984; 12 sets were collected to represent various moisture, weather, and seasonal conditions. Detailed notes were made on stream responses to changing moisture conditions in the watersheds, and on flow levels at each station in order to characterize the hydrology of the watersheds subjectively.

pH was determined potentiometrically at room temperature, within hours of collection. Cations were determined using atomic absorption spectrophotometry and anions were determined by automated colorimetric analysis. Bicarbonate alkalinity was determined by potentiometric titration without a fixed end point. Dissolved organic carbon (DOC) was determined by persulfate digestion to CO_2 measured by gas chromatography. Analytic methods used are described in more detail by Buso and others (1984).

Weathering Contributions

Rock types present in each watershed were determined by field reconnaissance and soil excavations. To relate rock types to stream chemistry, a simple method was devised to show the relative importance of chemical weathering. Ion availability in the watersheds is influenced by soil exchange properties, but is ultimately determined by chemical weathering of geologic materials. Samples of fresh rock were collected, crushed with a hammer, and sorted with wire sieves; 200 g of fragments that passed a 13-mm mesh but not a 6-mm mesh sieve were rinsed with distilled water to remove dust, then placed with 500 ml of distilled water in a capped, 1-liter, acid-washed, polyethylene bottle. After 27 days, the solutions were decanted into 500-ml, acid-washed, polyethylene bottles and analyzed the same way as stream samples.

The tests were conducted for 16 specimens chosen to represent the variety of rocks present. In addition, three rocks from the watershed of nearby Upper Hall Pond were included to test how known sulfidic rocks behave in the experiments. Several checks were used: (a) a control bottle containing only distilled water was maintained and analyzed, (b) the experiment was duplicated to check for reproducibility, and (c) analyses were duplicated after two months to check the stability of the solutions. The experiments provided a relative measure of the types of chemical weathering undergone by the various rock types.

Mineral Identification

Standard petrographic thin sections were prepared for eight of the rocks involved in the weathering experiments. The mineral composition of these rocks was estimated, and the rocks described. All other rocks were examined and described with the aid of a 10× hand lens. Mineralogic data were combined with the results of the weathering experiments to compare against differences in stream and pond chemistry.

Results

Stream Chemistry

Figure 1 characterizes the stream chemistry by showing average concentrations of selected ions for each site plotted against elevation. Although averages are plotted on these figures, the differences between sites were consistent on each sampling date. The chemistry of the upper portion of Falls Brook (Johnson and others 1981) is also plotted for comparison. Falls Brook is located within 20 km of our study watersheds.

For all parameters measured, the chemistry of Black Inlet is distinct from that of the Cone Inlet. Black Inlet is higher in basic cations and SiO_2. Cone Inlet is higher in H^+ and SO_4^{2-}. Black Inlet has appreciable alkalinity, while HCO_3^- alkalinity is nonexistent at the pH of Cone Inlet. Falls Brook lies intermediate

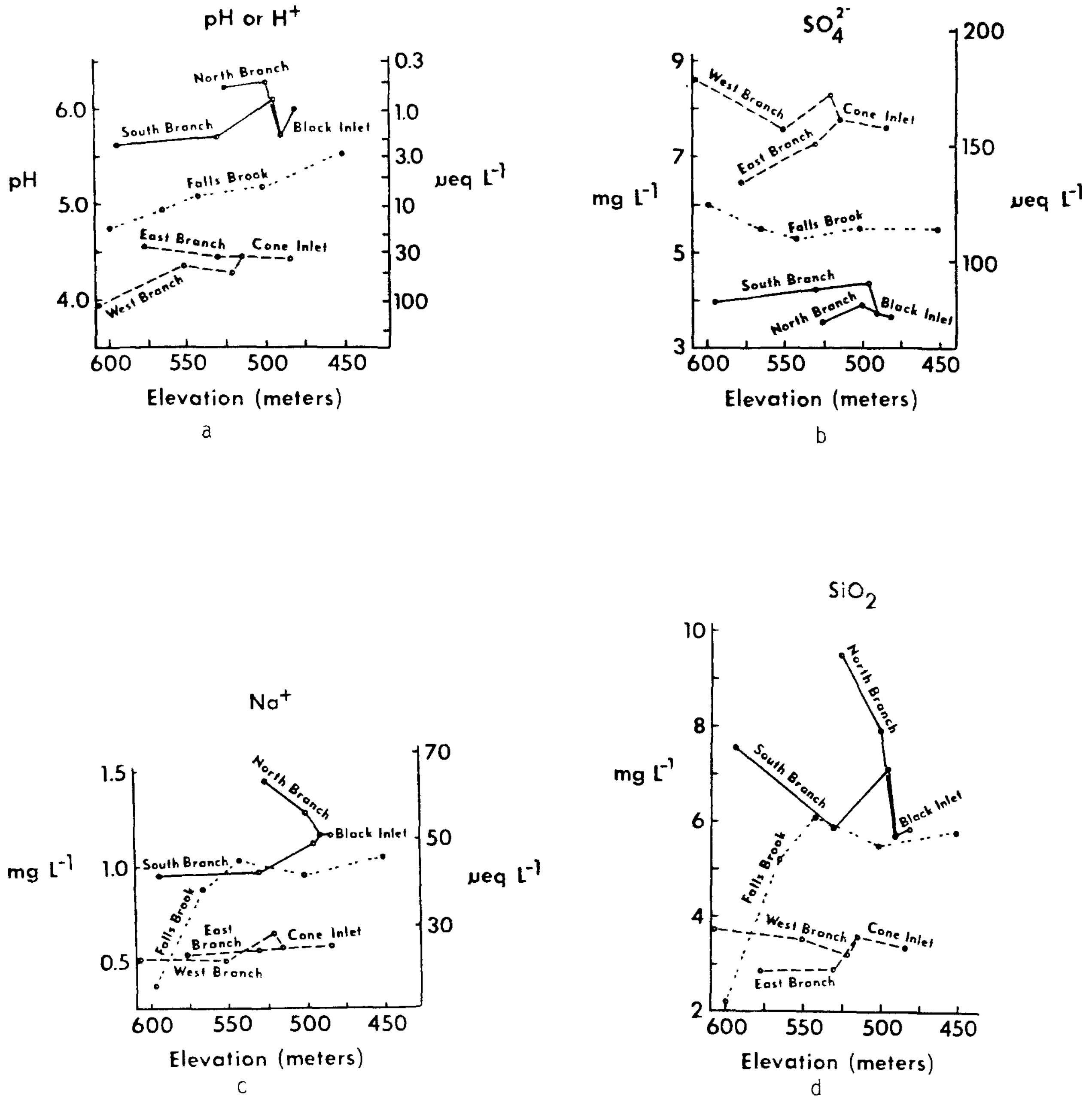

Figure 1. Streamwater concentrations for H^+ (a), SO_4^{2-} (b), Na^+ (c), and SiO_2 (d). Concentrations represent the mean of 12 sampling dates. Falls Brook data (Johnson and others 1981) represent the mean of 36 sampling dates. Add 215 m for Falls Brook elevations.

to Black and Cone on all parameters except for Mg^{2+}, which is comparable to Black.

Hydrology

The most significant point about the hydrology of Black Pond watershed is that the springs and streams were perennial during the entire summer of 1983. During this period, which included several months with below average rainfall (USDA Forest Service unpublished data), many area streams, some draining much larger watersheds, ceased flowing. The springs occur high in the Black Pond watershed, indicating that groundwater storage, possibly from outside the topographic divide, is likely contributing to streamflow. The slope above the watershed divide continues up to the summit of nearby Mount Flume at 1319 m. Subsurface flows may not be bound by the same features that define topographic watersheds in this area.

In contrast to Black Pond, a streamflow in the Cone Pond inlet system showed quick response to rainfall and snowmelt. All streams in the Cone Pond watershed stopped flowing in early June and did not flow again until a large storm in mid-August. Flow from this storm peaked quickly and streams were again dry until another storm in late October. Thereafter, streamflow was maintained through the rest of the study.

Geology

The till in both watersheds is loose and sandy. At Black, most of the recognizable rock fragments and abundant large surface boulders are Osceola Granite. Lafayette Granite Porphyry derived from the Franconia Ridge was found in minor amounts. Many other rock types, including prophyries, calcsilicate gneiss, camptonite, and slate, were also present in trace amounts.

The bedrock at Cone Pond watershed is largely metasedimentary quartz schist and mica schist. These two rock types are interbedded in layers of varying thickness. These rocks have been mapped as Littleton Formation (Billings 1956). Small 2-m pods of pegmatite are scattered through the metamorphic rocks. Also, a narrow vertical dike of basalt cuts through the central portion of the watershed. Other rock types were rarely observed in the till.

Mineralogy

Compositions of typical rocks from each watershed are shown in Table 2. Quartz is a major constituent of the rocks from Cone Pond watershed. Micas are also abundant. Muscovite and chlorite, which are more resistant to weathering than biotite (Carroll 1970), are more abundant. The rocks from Black Pond watershed also contain a large amount of quartz. However, there are large percentages of a variety of feldspars. Pyroxene, olivine, and a variety of amphiboles were also identified in rocks at Black. Sulfides are present in minor amounts in the metasedimentary rocks of Cone, and some of the prophyritic rocks from Black. Quartz and micas, the dominant minerals at Cone, are relatively stable in the weathering environment, whereas feldspars and amphiboles, common in the rocks at Black, are relatively less stable and thus good sources of basic cations (Carroll 1970).

Weathering Contributions

Solution chemistry for the artificial weathering experiment is summarized in Table 3. The solutions from Black contained high amounts of at least two basic cations. Of the six samples from Black, only one contained moderate SO_4^{2-}, and two samples had very high pH. Three samples were high in SiO_2. Alkalinity was much higher from Black samples compared to those from Cone.

None of the rocks tested from Cone yielded an appreciable amount of Ca^{2+}, Mg^{2+}, or Na^+. Only two specimens showed moderate amounts of SO_4^{2-}, and both were accompanied by higher H^+.

Samples of three rocks from the watershed of nearby Upper Hall Pond were tested to evaluate the behavior of known sulfidic rocks in the weathering experiments. Sulfate as high as 24,000 μeg liter^{-1} and hydrogen ion as high as 1050 μeg liter^{-1} were measured in these solutions. High amounts of basic cations were also observed. Although these rocks are part of the same Littleton Formation as the Cone samples, they obviously weathered differently.

Discussion

Stream Chemistry

Stream chemistry in the Black Inlet does not show the expected elevational gradient shown by nearby Falls Brook on which the acid neutralization model of Johnson and others (1981) was based. However, the chemistry of Black Inlet may still be explained by the same concepts which explain elevational gradients at Falls Brook—those of residence time and position along the stream channel. Since groundwater appears to be introduced primarily in the uppermost sections of Black Inlet, this stream is in effect a "Falls Brook in reverse."

Groundwater would have a relatively long path length, allowing longer reaction time between water and minerals. As pointed out by Johnson and others (1981), long path length implies more replacement of acidic cations by basic cations. Hydrologic response during our study strongly suggests that the streams are fed by a slow, steady supply of groundwater. All springs at Black occur high in the watershed. Thus, streamwater in the uppermost reaches has greater proportions of groundwater than lower reaches, where streamwater becomes diluted with surface and shallow subsurface flow. In turn, streamflow at lower elevation is characterized by higher H^+ and SO_4^{2-}, and lower basic cations and SiO_2. Chemical gradients support this thesis as concentrations of Ca^{2+}, Na^+, K^+, HCO_3^-, and SiO_2, the products of chemical weathering, all decrease in a downstream direction. Sulfate and DOC were observed to increase in a downstream direction. These gradients occurred during both baseflow and stormflow, but were more pronounced during stormflow.

Table 2. Estimated modes of rock samples in percent.

Mineral	Black Pond				Cone Pond				Stability	Weathering products
Sample no.	2	3	4	5	7	8	10	13		
Quartz	27	50	52		65	50	68	22	High	
Plagioclase			2	40					Mod. low	
Perthite	66	9	20						Mod. low	Sources of Ca^{2+}, Na^{+}, K^{+}
Orthoclase		34	tr						Moderate	
Biotite					15	tr	tr	9	Moderate	
Chlorite						20	10	13	Moderate	Sources of K^{+}
Muscovite					15	20	18	53	Mod. high	
Pyroxene			2	14					Low	
Arfvedsonite	7								Mod. low	
Kaersutite				25					Mod. low	Sources of Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}
Hornblende		6	15						Mod. low	
Olivine				6					Low	
Garnet		tr						tr	High	
Apatite			tr						Mod. low	Source of Ca^{2+}
Oxides	tr	1	7	10	5	8	3	1	High	
Sulfides			2	5	tr	2	1	2	Low	Source of H^{+}, SO_4^{2-}
Total	100	100	100	100	100	100	100	100		

tr, trace. Stability in the weathering environment, and weathering products after Carroll (1970).

Table 3. Weathering experiment solution chemistry.

Sample	Rock type	Abundance	H^{+}	Ca^{2+}	Mg^{2+}	Na^{+}	K^{+}	SO_4^{2-}	HCO_3^{-}	SiO_2
				BLACK POND WATERSHED						
1	Granite	Trace	2.2	0	0	8	6	24	<10	2.3
2	Granite	Major	1.2	100	12	19	70	8	80	9.4
3	Granite porphyry	Minor	1.0	80	3	21	28	17	NA	3.2
4	Granite porphyry	Trace	0.4	160	24	7	18	67	95	9.3
5	Camptonite	Trace	0.0	664	43	36	54	29	675	2.4
6	Calc silicate gneiss	Trace	0.2	190	10	15	16	29	135	7.4
				CONE POND WATERSHED						
7	Quartz schist	Major	2.8	0	2	2	14	28	<10	2.1
8	Quartz schist	Minor	3.5	0	2	4	14	27	<10	2.7
9	Quartz schist	Major	2.3	0	1	1	15	18	<10	0.8
10	Quartz schist	Major	10.0	5	5	1	16	69	0	1.5
11	Mica schist	Major	1.6	0	1	3	17	17	<10	1.5
12	Mica schist	Major	0.8	5	2	5	10	18	<10	3.0
13	Mica schist	Major	5.2	10	12	4	32	92	0	1.8
14	Granite pegmatite	Minor	1.7	0	0	3	5	23	<10	0.8
15	Basalt	Trace	0.5	20	16	10	32	12	80	3.6
16	Quartz monzonite	Trace	0.9	5	1	3	13	8	20	1.2
				UPPER HALL POND WATERSHED						
17	Quartz schist	Minor	13	206	92	3	55	458	0	3.1
18	Pyrite mica schist	Major	1050	110	574	3	18	24000	0	36
19	Volcanic breccia	Trace	0.1	857	44	15	18	12	780	1.7
				CONTROL						
20	Control		1.1	0	0	0	0	0	0	0

At Cone Inlet, no consistent trends were noted on an elevational gradient. Again, the gradients observed at Falls Brook were not found. However, water with longer residence time in the soil and geologic materials is never introduced to Cone Inlet because of shallow soils and expansive bedrock outcrops over the entire length of the stream. The last section of the inlet, below the junction of the branches, flows through a rocky gorge and over waterfalls, which prohibit the introduction of subsurface flows. The result is that the entire length of the Cone Inlet resembles the uppermost portion of Falls Brook.

The hydrology of Cone Inlet is consistent with the above explanation. The streams are quick to respond to changing moisture conditions in the watershed, indicating that most streamflow arises from the soil mantle, and not from groundwater. The lack of feldspars and ferromagnesian minerals coupled with the dominance of quartz and muscovite results in minimal chemical weathering.

The same factors which contribute to the patterns noted in stream chemistry within the watersheds are responsible for differences noted between the two watersheds. Waters at Black are enriched in basic cations, SiO_2 and HCO_3^-, compared to those at Cone. Overall, water at Black is subject to longer path lengths and residence time within the watershed than at Cone. Differences in soils, outcrop distribution, and runoff timing all support this conclusion. Also, the rocks at Black are more reactive in terms of basic cations and alkalinity than those at Cone. This results in a larger buffering reservoir at Black.

Weathering Contributions

In the weathering experiment, rocks from Black Pond watershed produced an average of ten times the equivalent of basic cations as rocks from Cone. Sulfate production seems to be minor in the rocks of both watersheds. The rocks of Upper Hall Pond watershed weathered differently, producing large amounts of basic cations and SO_4^{2-}.

Sulfate levels in the two watersheds are not completely explained by this study. Black Inlet has low SO_4^{2-}, at 3–4 mg liter^{-1}, while Cone Inlet has remarkably high SO_4^{2-}, at 7–15 mg liter^{-1}, compared to other small streams in the area (Johnson and others 1981, Martin 1979, Fisher and others 1968). The inlet to Upper Hall Pond was sampled three times and found to have about average SO_4^{2-} for the area, at 6–7 mg liter^{-1}. Mean SO_4^{2-} in precipitation at the nearby Hubbard Brook Experimental Forest is 2.9 mg liter^{-1} (Likens and others 1977).

The rocks of Black Pond watershed are potentially minor sources of SO_4^{2-} (Table 3). One rock type of minor abundance produced modest amounts of SO_4^{2-}, while others produced trace amounts. The major rocks of Cone Pond watershed as a group produced relatively small amounts of SO_4^{2-} in the weathering experiments. On the other hand, rocks from Upper Hall Pond watershed demonstrate that known sulfide-bearing rocks are capable of producing large amounts of SO_4^{2-}.

These results indicate that the mineralogy of these watersheds is apparently not the controlling factor in determining SO_4^- levels in streamwater. Sulfidic materials exposed to cycling water may already be completely weathered of sulfides or coated by weathering crusts that inhibit further weathering. Other factors such as adsorption on soils, oxidation reduction in wetlands, uptake by vegetation, and differential entrapment of dryfall may be influencing SO_4^{2-} cycling as well. Temporal patterns and multistation sampling suggest that SO_4^{2-} accumulates in Cone Pond watershed peatlands during dry periods and is flushed out of the system in the first flow following heavy rains.

Implications for Acidification

Likens and others (1977) show that SO_4^{2-} is the dominant anion in stream water in the northeast. On a geologic time scale, however, the H_2CO_3 system is considered to be dominant over the H_2SO_4 weathering system (Johnson and others 1972). A change from H_2CO_3 to H_2SO_4 would have met little buffering from the mineral materials at Cone Pond watershed. Also, path lengths and residence time of water are relatively short, so the opportunity for buffering reactions is limited. Based on these factors, acidification by H_2SO_4 would have proceeded relatively unhindered.

Ford (1984) has suggested that Cone Pond has been acidic for thousands of years based on the diatom assemblages observed in sediment cores. These data, along with prevalence of sphagnum peatlands on the inlet streams, and the inert character of the underlying geology seem to indicate that Cone Pond may have been a brown water acid pond which was converted to a clear-water acid pond because of recent sulfate deposition. Recent conversion may have occurred as humic compounds flocculated due to increasing Al and SO_4^{2-}, similar to effects documented for lakes in Norway (Davis and others 1985).

Increased H_2SO_4 inputs to Black Pond watershed would be buffered by reactions with the minerals in the rocks, till, and soil, as well as by the groundwater component of streamflow. How long this buffering would last in the event of increased H_2SO_4 levels is unknown. Buffering is present, but total dissolved solids are low compared to some other watersheds in the region (Johnson and Reynolds 1977).

Our study indicates that identification of key minerals, each of which may be present in a large variety of rock types, is especially important in characterizing watershed buffer capacity. Olivines, pyroxenes, and amphiboles are important sources of basic cations. On the other hand, minerals such as quartz and muscovite are relatively inert to chemical weathering. Weathering of sulfides yields strong acids. Therefore, weathering of specific minerals may add to or subtract from the acidity of solutions. In addition, weathering of sulfides may generate enough acidity so that otherwise stable minerals are dissolved.

The characterization of a watershed's buffering capacity or aquatic chemistry based on rock types as suggested by Norton (1980) may be misleading. In this study, we found a granitic watershed much better buffered than a metasedimentary watershed. This is just the opposite of what would have predicted by the bedrock susceptibility scheme. It may be more appropriate to characterize a watershed by the minerals present than by general rock types. The granite from Black Pond watershed contains the reactive mineral arfvedsonite. This granite has a very different influence on water chemistry than would a granite with micas instead of amphiboles. Further, our experiments suggest that complex units, such as the Littleton Formation, can have widely different weathering characteristics. It may be possible to refine susceptibility classifications by rating areas based on the presence of certain minerals rather than on general rock types.

The results of this study also point out that the regional approach to classifying the sensitivity of streams and lakes to acidification (Omernik and Powers 1982) is often inappropriate for individual streams and ponds. Factors not easily obtainable from regional maps, such as stream hydrology, contributions from groundwater and springs, wetlands, mineralogy, and soil depth and permeability, have strong influences on stream and pond chemistry. This results in each stream and pond having a unique character determined by the configuration and components of its own watershed.

Conclusion

Our study suggests several relatively simple observations and experiments that aquatic managers might apply to assess susceptibility to acid deposition. Monitoring of stream chemistry along an elevational gradient yields data that can reveal the influence of geologic weathering and groundwater inputs to the downstream evolution of stream chemistry. Whereas monitoring of a stream at one location may be sufficient to characterize it as "acidic" or "poorly buffered" at that site, the management of a stream for uses such as water supply or fisheries may depend on the entire reach. Multistation sampling should be used to determine the chemistry of a stream and the causes of chemical gradients.

Observations of stream hydrology may further refine understanding of the dynamics of a particular stream. Stream gauging and groundwater monitoring programs may be used to characterize response to changing moisture conditions and locations and amounts of groundwater discharge. These factors are important determinants of path length of water within the watershed, and therefore contact with buffering or acidifying components of soils and geologic materials.

Artificial weathering experiments are an inexpensive, simple technique that may be used by aquatic managers to assess the potential influence of bedrock on stream chemistry in watersheds where fresh rock samples may be obtained. A knowledge of mineral identification is not necessary, only the capability of conducting or obtaining chemical analyses of the sample solutions. Rock types which are inert or easily weathered may be identified. In addition, easily weathered rocks may be characterized as sources of basic or acidic weathering products.

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