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Sensitivity to acidification of subalpine ponds and lakes in north-western Colorado

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Abstract:

Although acidifying deposition in western North America is lower than in many parts of the world, many high-elevation ecosystems there are extremely sensitive to acidification. Previous studies determined that the Mount Zirkel Wilderness Area (MZWA) has the most acidic snowpack and aquatic ecosystems that are among the most sensitive in the region. In this study, spatial and temporal variability of ponds and lakes in and near the MZWA were examined to determine their sensitivity to acidification and the effects of acidic deposition during and after snowmelt. Within the areas identified as sensitive to acidification based on bedrock types, there was substantial variability in acid-neutralizing capacity (ANC), which was related to differences in hydrological flowpaths that control delivery of weathering products to surface waters. Geological and topographic maps were of limited use in predicting acid sensitivity because their spatial resolution was not fine enough to capture the variability of these attributes for lakes and ponds with small catchment areas. Many of the lakes are sensitive to acidification (summer and autumn ANC < 100 μeq L^{-1}), but none of them appeared to be threatened immediately by episodic or chronic acidification. In contrast, 22 ponds had minimum ANC < 30 μeq L^{-1}, indicating that they are extremely sensitive to acidic deposition and could be damaged by episodic acidification, although net acidity (ANC < 0) was not measured in any of the ponds during the study. The lowest measured pH value was 5·4, and pH generally remained less than 6·0 throughout early summer in the most sensitive ponds, indicating that biological effects of acidification are possible at levels of atmospheric deposition that occurred during the study. The aquatic chemistry of lakes was dominated by atmospheric deposition and biogeochemical processes in soils and shallow ground water, whereas the aquatic chemistry of ponds was also affected by organic acids and biogeochemical processes in the water column and at the sediment–water interface. These results indicate that conceptual and mechanistic acidification models that have been developed for lakes and streams may be inadequate for predicting acidification in less-understood systems such as ponds.

KEY WORDS atmospheric deposition; lakes; ponds; acidification

INTRODUCTION

Regional surveys have shown that the snowpack in and near the Mount Zirkel Wilderness Area (MZWA) is the most acidic in western North America (Turk and Campbell, 1997; Turk *et al.*, 2001). This is supported by precipitation chemistry data from nearby Buffalo Pass, where annual volume-weighted-mean field pH values ranged from 4·6 to 5·0 during 1984–1999 (NADP/NTN, 2001). Emissions of sulphur dioxide and particulates from two coal-fired power plants in the Yampa Valley upwind of the MZWA were found to contribute to visibility impairment in the MZWA (Watson *et al.*, 1996); power plant emissions of sulphur and nitrogen oxides also contribute to the acidity of the precipitation. One of the Yampa Valley power plants recently installed emissions controls designed to reduce sulphur dioxide emissions by 85% and nitrogen oxides by

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50%, and the other plant is scheduled to implement similar controls by 2004 (D.W. Ely, Colorado Department of Public Health and Environment, personal communication), so the acidity of precipitation in the MZWA is expected to decrease in future years. Thus, the MZWA provides a unique opportunity to determine (i) the extent of impacts on ecosystems that have received the most acidic deposition in the region during recent years, and (ii) the response of deposition and ecosystems to reduced emissions during future years.

Sensitivity of surface waters to acidification is determined in most undisturbed systems by the availability of weathering products (base cations and acid-neutralizing capacity (ANC)) to neutralize acidic components of atmospheric deposition (primarily H_2SO_4 and HNO_3). The availability of weathering products in watersheds is controlled by (i) supply, i.e., the quantity and replenishment rate of weathering products stored in soil and ground water, and (ii) transport, i.e., the rate that the weathering products are delivered to surface waters, which is determined by hydrological flowpaths and hydrological residence times.

Many aquatic ecosystems in the Rocky Mountains are vulnerable to the effects of acidification from atmospheric deposition because they have thin soils, relatively unreactive bedrock, and hydrological flowpaths that allow little neutralization of acids in snow and rain (Turk and Spahr, 1991). A survey in 1983 indicated that about one-half of the lakes in the MZWA had ANC of less than 100 μeq L^{-1} (Turk and Campbell, 1987). The acid-base chemistry of selected lakes in the MZWA has been monitored during summer since 1985 (Turk *et al.*, 1993), but there have been no surveys or long-term monitoring of vernal ponds. Nor have data been collected during the snowmelt period, when lakes and ponds are most vulnerable to episodic acidification from the pulse of acidic meltwater from the snowpack.

Few studies have been done on the effects of acidic deposition on biological resources in these sensitive high-elevation Rocky Mountain ecosystems. Some studies suggested that the tiger salamander (*Ambystoma tigrinum*) is sensitive to acidity at levels that might be found in snowmelt and rainfall in the MZWA (Harte and Hoffman, 1989; Kiesecker, 1996; Turk and Campbell, 1997). Corn and Vertucci (1992) concluded that acidic deposition was not likely a regionwide factor in amphibian declines in the Rocky Mountains; however, the Rabbit Ears Pass site near Dumont Lake was described as the most likely out of 57 sites inspected to be at risk for chronic acidification (sulphate deposition >10 kg ha^{-1} $year^{-1}$ and ANC < 200 μeq L^{-1}).

The objectives of this study were to (i) determine the acid-base chemistry of ponds and compare their sensitivity to acidification to that of lakes, and (ii) determine the acidity of lakes and ponds during and after snowmelt, when they are most vulnerable to episodic acidification. A companion study evaluated whether episodic or chronic acidification is affecting reproductive success of amphibians (Muths *et al.*, 2003).

STUDY AREA

The study area is in and immediately south of the Mount Zirkel Wilderness in the Park Range of north-western Colorado (Figure 1). The study sites at Dumont Lake, Buffalo Pass and Mount Ethel straddle the Continental Divide and are characterized by a cold, continental climate with snow cover for more than 6 months of the year. Vegetation is patchy subalpine spruce–fir forest intermingled with grassy meadows and wetlands.

The Dumont study site is an area of numerous small ephemeral ponds near Dumont Lake (elevation about 2900 m) on the east side of Rabbit Ears Pass. Ponds are located within 3 km of U.S. Highway 40 and are accessible year-round. Bedrock geology is primarily volcanic (Snyder, 1980b) and soils are poorly developed. Ponds are present in a variety of landscape positions including drainages, closed topographic depressions, tops of knolls and toes of hillslopes. The Dumont ponds are typically covered by 1–3 m of snow by spring and become ice-free in late May. Weather in late May and early June is variable and may include warm sunny days, afternoon thunderstorms, or extended periods of rain or snow. These extremes in weather conditions can cause the ponds at this site to become ice-free at different times and warm slowly. Snowmelt is interrupted during stormy periods, and cold temperatures can refreeze pond surfaces.

Buffalo Pass lies on the southern boundary of the MZWA and is accessible by snowmobile during winter and by automobile from July through to October. Most of the ponds at the Buffalo Pass study site are at an elevation of about 3200 m and are located 1–4 km inside the wilderness boundary. Bedrock geology is

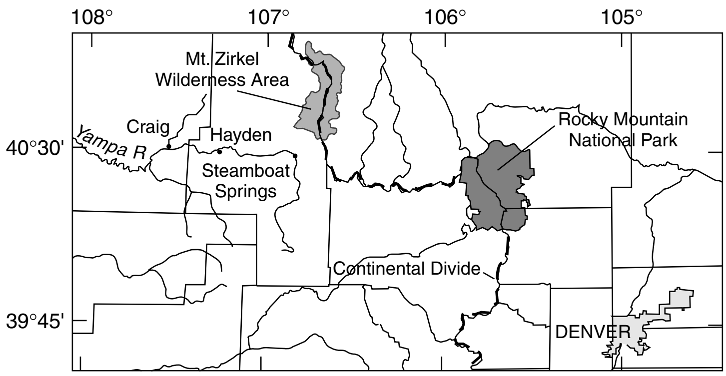

Figure 1. Location map

primarily granitic (Snyder, 1980a) and previous studies identified lakes in the area as sensitive to atmospheric deposition (Turk and Campbell, 1987). The ponds at this site are located in a variety of landscape positions, including some perched along ridges or cliffs that appear to be disconnected from any groundwater flow system. Buffalo Pass ponds are typically covered by 2–4 m of snow in winter and spring and become ice-free in late June. This is the time of year with maximum solar radiation, warm temperatures and relatively few afternoon thunderstorms or extended stormy periods. Most ponds near Buffalo Pass become ice-free at about the same time and warm rapidly, with water temperatures reaching 10 °C within 1 week of becoming ice-free.

The Mount Ethel site lies within the MZWA approximately 10 km north of Buffalo Pass site at an elevation of 3340 m. Climate, geology and landscape position of the ponds in this area are similar to ponds at the Buffalo Pass site.

The chemical composition and sensitivity to acidification of lakes near the Buffalo Pass and Mount Ethel sites are described in Turk and Campbell (1987). Most of the lakes have inflows and outflows and would be classified as drainage lakes, although some such as Summit Lake have little surface inflow and only seasonal outflow and are dominated by seepage.

For the purpose of this study, ponds are defined as having maximum depth of less than 2 m. Some had a maximum depth of as little as 0·5 m. The diameter or distance across the longest axis of the ponds was usually less than 100 m. None supported fish populations. Note that the name 'George Lake' as found on topographic maps was actually a pond by this definition. In contrast, most lakes had a maximum depth much greater than 2 m, a diameter of 100 m or greater, and many supported fish. By late winter, lakes in the region typically have a few meters of slush and thin ice layers covering unfrozen water, whereas ponds that do not dry out by autumn have layers of slush and ice to their full depth.

METHODS

The study was designed to determine spatial and temporal variability in chemical composition of ponds and lakes in the MZWA. During 1998, synoptic sampling was done to identify ponds suitable for more intensive study in 1999. The intensive study focused on six ponds at each study site—three that were among the lowest ANC, and three that were relatively insensitive to acidification. Specific ponds were chosen also for their suitability for the companion study to determine acidification effects on amphibians (Muths *et al*., 2003). The Dumont study site was chosen because of the earlier research conducted there on acidification and amphibians (Kiesecker, 1991; Kiesecker, 1996). The Buffalo Pass study site was chosen because of the lakes monitoring going on there (Turk and Campbell, 1987; Turk *et al*., 1993) and because it was within the MZWA, a Class 1 Area protected from impacts of air quality degradation by the Clean Air Act Amendments.

In 1998, numerous ponds and lakes at the Dumont and Buffalo Pass sites were sampled one or more times. Most ponds that could be identified on 7·5′ topographic maps, and many additional ponds identified in the field, were sampled at least once. Temporal changes during early summer also were monitored in selected ponds that represented a range of sensitivities to acidification. One hundred and seventy samples were collected from 39 ponds and Dumont Lake at the Dumont site. One hundred and fifty-nine samples were collected from 30 ponds and 16 lakes at the Buffalo Pass site.

In 1999, more intensive sampling was done at six ponds each at the Dumont and Buffalo Pass sites. The ponds were selected based on the design of the amphibian experiment (Muths *et al*., 2003), which required a cluster of three low-ANC ponds and three high-ANC ponds at each site. The low-ANC ponds were chosen carefully and represent ponds in the lower 10th percentile of ANC values, whereas the high-ANC ponds had ANC values above the median but not always at the extreme. All of the high-ANC ponds were relatively insensitive to acidification and thus the six ponds at each site represent a range of response to acidification that would be expected at each study site. Samples were collected every 2 days at these sites during early summer of 1999. Additional samples were collected occasionally from other ponds at Buffalo Pass and once during August of 1999 from the Mount Ethel site. Earliest samples in both years were collected prior to or a few days after each pond became ice-free.

Monitoring of a subset of the intensively monitored sites at Buffalo Pass has continued through 2002. The more recent data were used to determine the relationship between anion deficit and DOC, which was not measured during 1998 or 1999.

Samples were collected manually from the outlets of ponds and lakes when they were flowing, or from shoreline locations with minimal vegetation. Samples were refrigerated, and aliquots were divided and filtered within 24 h.

Specific conductance, pH and ANC were measured in the laboratory on unfiltered, unpreserved, air-equilibrated samples. Gran titration for ANC was performed to pH 3·0 on a Radiometer Automated Low Ionic Strength Titrator (use of trade name is for information only, and does not constitute endorsement or approval by the U.S. Geological Survey), laboratory pH was measured by this instrument prior to titration. Filtered (0·45 μm), refrigerated aliquots were used to analyse K, Na, NH_4, Cl, NO_3 and SO_4 by ion chromatography. Filtered aliquots preserved with nitric acid (pH less than 2) were used to analyse Ca, Mg and SiO_2 by inductively coupled plasma spectroscopy. Dissolved organic carbon (DOC) was measured by UV-promoted persulfate oxidation and infrared spectrometry. Detection limits were less than 0·3 mg L^{-1} for DOC, 3 μmol L^{-1} for SiO_2 and 1 μmol L^{-1} for all other constituents.

Quality assurance included collection of field blanks and duplicates (approximately 10% of all samples). Sample data were checked for outliers based on charge balance, measured versus calculated specific conductance, and time-series plots. Outliers were reanalysed to confirm or correct results.

Data on atmospheric deposition came from the National Atmospheric Deposition Network/National Trends Network (NADP/NTN) site at Buffalo Pass (NADP/NTN, 2001). Snowmelt acidity calculations were estimated based on snowpack acidity measured during the study, and meltwater amplification factors from an earlier study. The bulk snowpack acidity at peak accumulation of snow in April of each year of the study was calculated as the median of data from three nearby sites (Buffalo Pass, Hogan Peak and Dry Lake) (Turk *et al*., 2001). Meltwater amplification factors were calculated from snowmelt lysimeter data in 1995 at Loch Vale, Rocky Mountain National Park, located approximately100 km east of the sites in this study (Campbell, 1999). The meltwater amplification factor used to simulate the ionic pulse during snowmelt (Johannessen and Henriksen, 1978) is calculated as the acidity at a given time divided by the volume-weighted mean acidity of all meltwater samples.

RESULTS

Snowmelt acidity

The estimated acidity of snowmelt in the study area prior to neutralization by weathering products is presented in Figure 2. Calculated pH values ranged from 4·2 during early snowmelt to 5·5 later in the spring. Spatial and temporal variability in climate affect meltwater amplification factors—the meltwater amplification factors calculated from snowmelt measured in 1995 at Loch Vale in Rocky Mountain National Park were within the lower end of the range reported in other studies, and probably represent a conservative calculation of meltwater acidity for sites in this study.

Dumont ponds

Minimum ANC measured at each of the Dumont ponds in 1998 is presented in a frequency histogram (Figure 3). Values of ANC varied substantially between ponds within the Dumont site. Most ponds had minimum ANC of greater than 50 μeq L^{-1}, and some were greater than 200 μeq L^{-1}.

The six Dumont ponds that were monitored intensively in 1999 were divided into low-ANC and high-ANC groups for biological experiments described by Muths *et al.* (2003). The range of ANC in the Dumont low-ANC ponds had ANC that overlapped the range in the Buffalo Pass high-ANC ponds, thus none of the ponds at Dumont was very sensitive to acidification. In samples collected as ice melted in 1999, ANC in the Dumont ponds ranged from 100 to 130 μeq L^{-1} for high-ANC ponds and 20 to 90 μeq L^{-1} for the low-ANC ponds (Figure 4). Sum of base cations (SBC: $Ca^{+2} + Mg^{+2} + Na^{+} + K^{+}$) as ice melted ranged from 100 to 200 μeq L^{-1} for high-ANC ponds, and 50 to 130 for low-ANC ponds. Sum of acid anions (SAA: $SO_4{}^{-2} + NO_3{}^{-} + Cl^{-}$) was 10 to 30 μeq L^{-1} in all of the ponds. Concentrations of Cl^{-} and $SO_4{}^{-2}$ in the ponds were approximately equal to those in the snowpack (1–3 μeq L^{-1} and 8–15 μeq L^{-1}, respectively), indicating that watershed sources of these ions were not present to complicate effects of acidic deposition. Concentrations of $NO_3{}^{-}$ were substantially less in the ponds than in the snowpack (0–6 μeq L^{-1} in the ponds compared with 11–17 μeq L^{-1} in the snowpack), indicating biological uptake of nitrogen in terrestrial and/or aquatic ecosystems. Thus, the SAA in the ponds generally reflected atmospheric deposition with partial nitrate removal.

The timing and magnitude of the changes in chemical composition varied between individual ponds at the Dumont site. In the three high-ANC ponds and Dumont Pond 25, ANC decreased during snowmelt, then increased from early June through to the end of June. In Dumont Pond 11, SBC and ANC decreased through mid-June then increased slightly by the end of June. In Dumont Pond 14, SBC and ANC were relatively low when sampling began and increased steadily except for a slight decrease in late June. In all the ponds, SAA ranged from 13 to 26 μeq L^{-1} in the earliest samples and decreased to 5–10 μeq L^{-1} by late June. This decrease was probably caused by biological assimilation of sulphate and nitrate: SO_4 in all three ponds

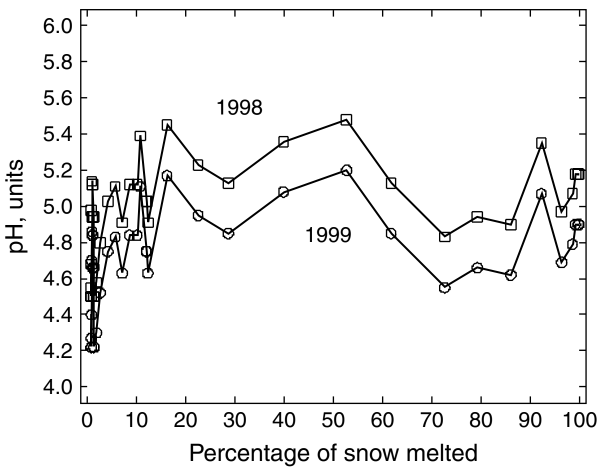

Figure 2. Predicted acidity, expressed as pH, of snowmelt in Mount Zirkel Wilderness Area in 1998 and 1999. Calculated as the product of the median bulk snowpack acidity for three nearby snowpack chemistry monitoring sites (Buffalo Pass, Hogan Peak and Dry Lake) and the amplification factors in the acid pulse of snowmelt measured at Loch Vale in Rocky Mountain National Park, Colorado, in 1995

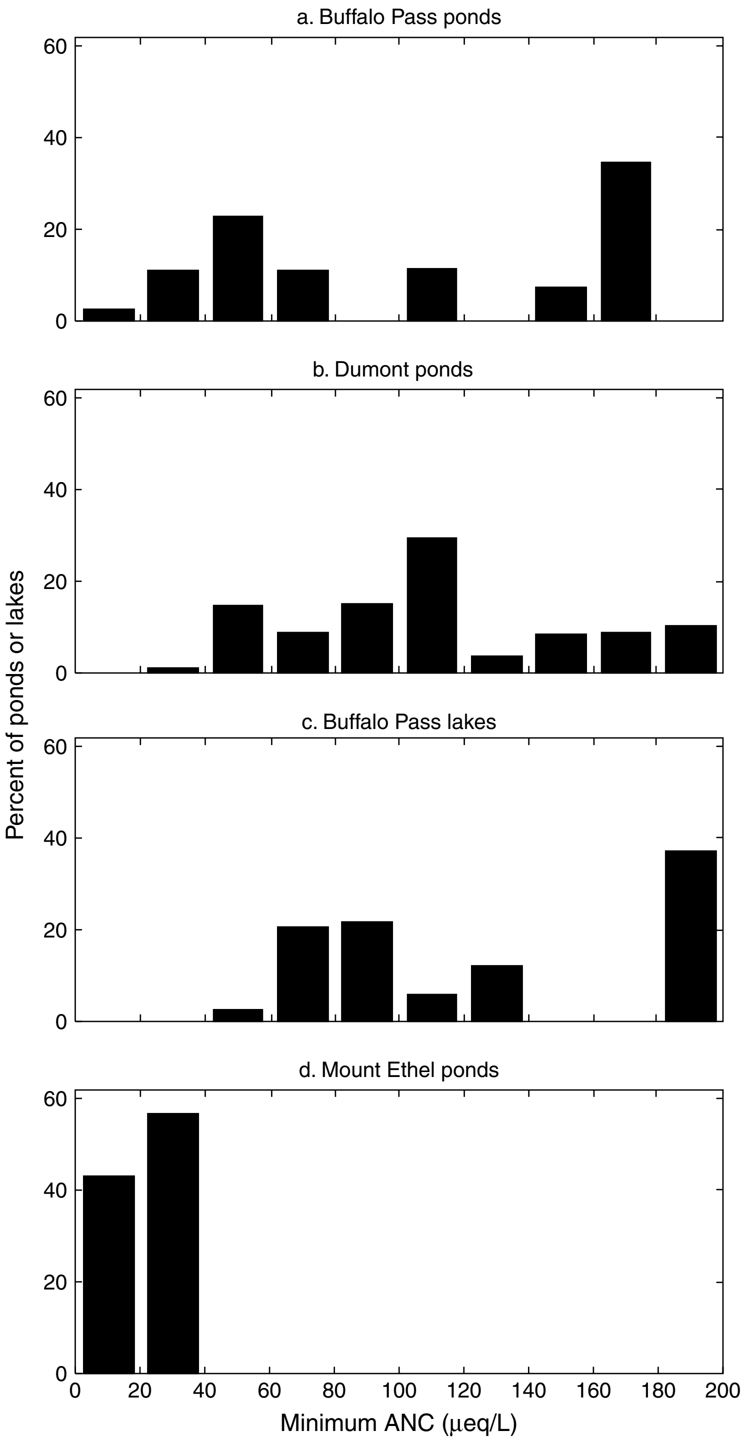

Figure 3. Frequency histogram of ANC (μeq L^{-1}). Data presented are minimum concentrations in 1999 for Mount Ethel ponds, and minimum concentrations in 1998 at other sites

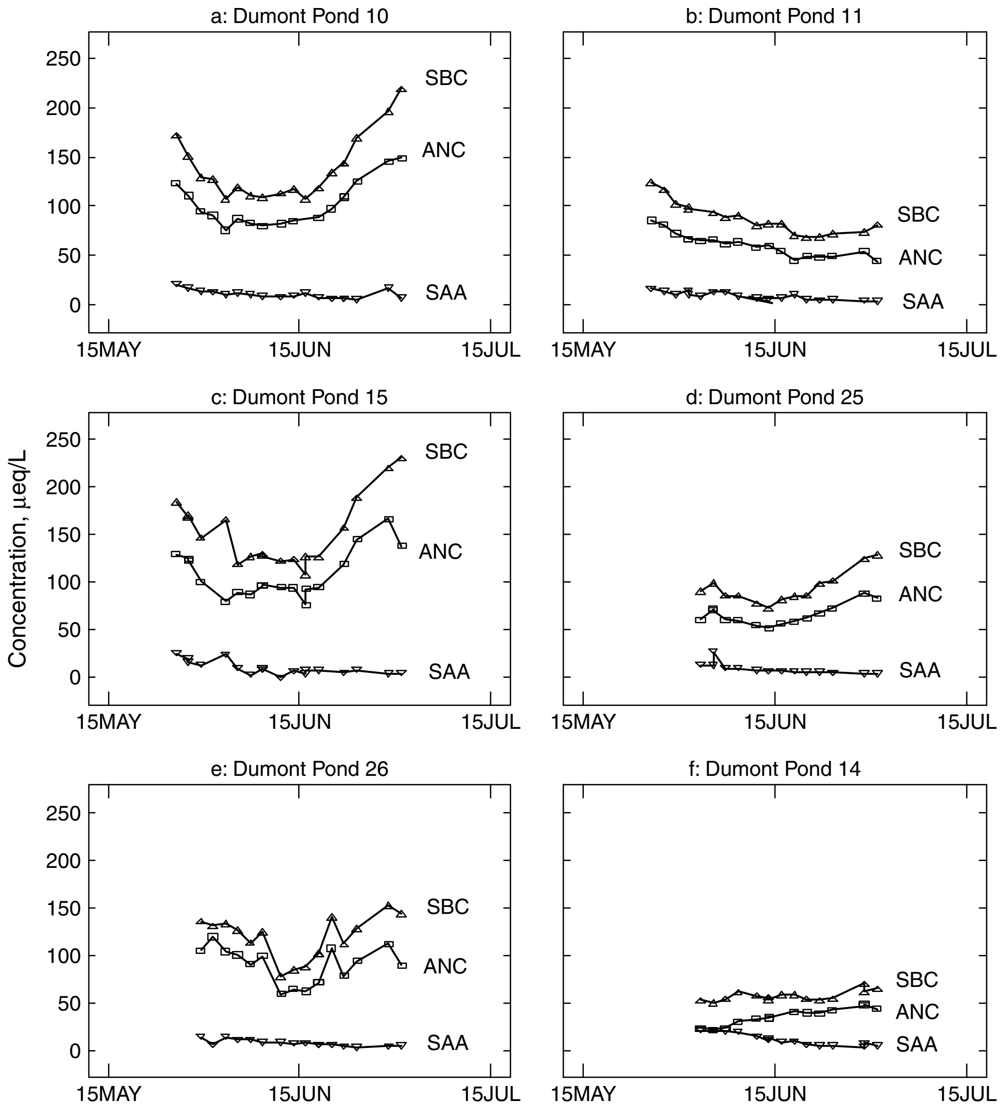

Figure 4. Sum of base cations (SBC), sum of acid anions (SAA) and acid-neutralizing capacity (ANC) for Dumont Ponds during 1999. Units are μeq L^{-1}. Left column is high-ANC class; right column is low-ANC class

decreased to less than 5 μeq L^{-1} and NO_3 decreased to less than the detection limit, whereas Cl did not change significantly.

Acid-neutralizing capacity was controlled by SBC in most of the Dumont ponds and was reduced only slightly by acid anions. The exception was Dumont Pond 14, where the seasonal decrease in SAA was comparable to the increase in SBC, resulting in an overall increase of ANC from 22 to 48 μeq L^{-1} during the sampling period.

Buffalo Pass ponds

Sensitivities to acidification varied greatly in the Buffalo Pass ponds (Figure 3). Some ponds were relatively insensitive, others had sensitivity comparable to that of nearby lakes (Figure 3 and Turk and

Campbell 1987), and some were among the most sensitive aquatic ecosystems identified in western North America.

Many of the ponds had ANC comparable to values reported for the Dumont ponds. Most ponds had minimum ANC of greater than 30 μeq L^{-1}, and some were greater than 100 μeq/l. However, 12 ponds had minimum ANC of less than 30 μeq L^{-1}, making them extremely sensitive to acidification. The lowest ANC measured was 3 μeq L^{-1} at Pond 98-18 on 6 August 1998 (Figure 5), and the lowest pH measured was 5·37 at Pond 98-14 on 18 July 1999.

Concentrations of ANC in the three high-ANC Buffalo Pass ponds were 40–60 μeq L^{-1} immediately after ice melted in 1999. Sum of base cations ranged from 70 to 90 μeq L^{-1}, and SAA was 10–20 μeq L^{-1}. Acid

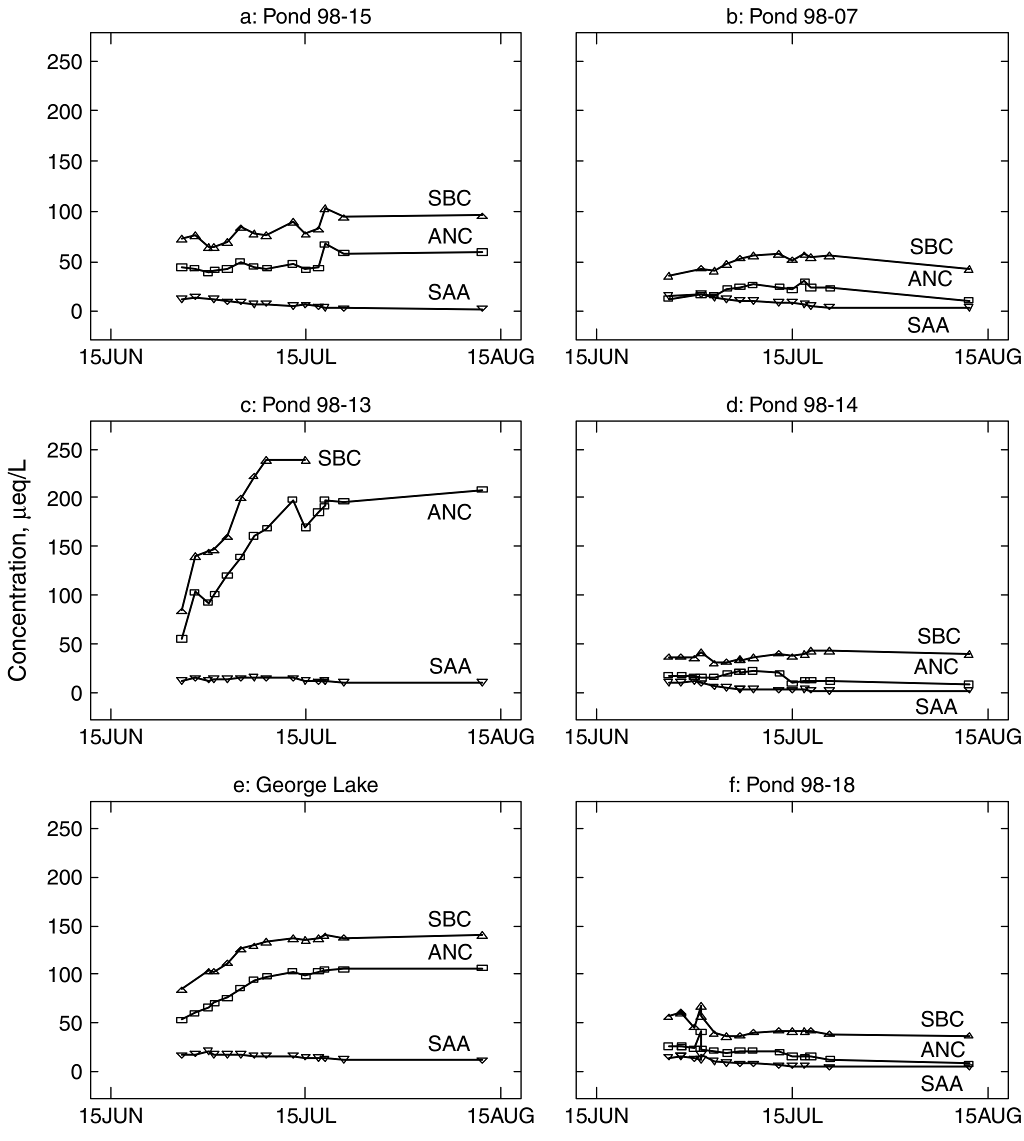

Figure 5. Sum of base cations (SBC), sum of acid anions (SAA) and acid-neutralizing capacity (ANC) for Buffalo Pass Ponds during 1999. Units are μeq L^{-1}. Left column is high-ANC class; right column is low-ANC class

anions reflected atmospheric deposition with partial nitrate removal, similar to the Dumont ponds. During the first 4 weeks of the ice-free period, SBC increased in all of the high-ANC Buffalo Pass ponds; the range in SBC for the three ponds on 21 July was 95–270 μeq L^{-1}. During this period, SAA decreased to less than 5 μeq L^{-1} in these ponds, similar to the Dumont ponds. The net result was an increase in ANC to 55–200 μeq L^{-1}. From mid-July to mid-August, concentrations of SBC, SAA and ANC changed little in the three ponds.

Concentrations of ANC in the low-ANC Buffalo Pass ponds were 10–20 μeq L^{-1} immediately after ice melted. Sum of base cations ranged from 30 to 60 μeq L^{-1}, and SAA was 10–20 μeq L^{-1}. As with the Dumont ponds, acid anions reflected atmospheric deposition with partial assimilation of sulphate and nitrate (Figure 6). During the first 4 weeks of the ice-free period (through to 21 July), SBC increased in Ponds 98-07 and 98-14, whereas it decreased in Pond 98-18. The range in SBC for the three ponds was 39–56 μeq L^{-1}. Sum of acid anions decreased to less than 5 μeq L^{-1} in all of the ponds. The net result was a decrease to 12–13 μeq L^{-1} ANC in Ponds 98-18 and 98-14 and an increase to 23 μeq L^{-1} ANC in Pond 98-07. By late summer (14 August), SBC had decreased slightly in all three of the low-ANC ponds, resulting in ANC of 8–11 μeq L^{-1} in all three ponds.

Mount Ethel ponds

The very low ANC values detected in some of the Buffalo Pass ponds raised the question of whether similar ponds might exist elsewhere in the MZWA. A number of relatively inaccessible ponds on a ridge near Mount Ethel were sampled once in August of 1999. The ANCs measured in these ponds are presented in a frequency histogram (Figure 3). All of these ponds had ANC of less than 40 μeq L^{-1}, and almost one-half of them had ANC of less than 20 μeq L^{-1}, even in midsummer well after the snowmelt pulse. These ponds are extremely sensitive to acidification.

Buffalo Pass lakes

Minimum ANC measured in 1998 at each of the lakes is presented in Figure 3. Most lakes had minimum ANC of 60–140 μeq L^{-1}, one was slightly less, and three lakes had ANC of greater than 180 μeq L^{-1}. These results are consistent with the broader survey of lakes in the region in 1983 (Turk and Campbell, 1987).

Of the Buffalo Pass lakes, Summit Lake was the most sensitive to acidification based on the 1998 survey, therefore it was intensively monitored along with the ponds at Buffalo Pass during 1999. A sample collected from the outlet stream on 15 June, after snowmelt had begun but when the lake was completely ice- and snow-covered, had an ANC of 54 μeq L^{-1} (Figure 7). By 26 June (the earliest sample date for most of the ponds at Buffalo Pass), the ANC at Summit Lake decreased to 46 μeq L^{-1}, but 4 days later ANC was 77 μeq L^{-1}, and it remained between 60 and 78 μeq L^{-1} through to mid-September. Sum of base cations

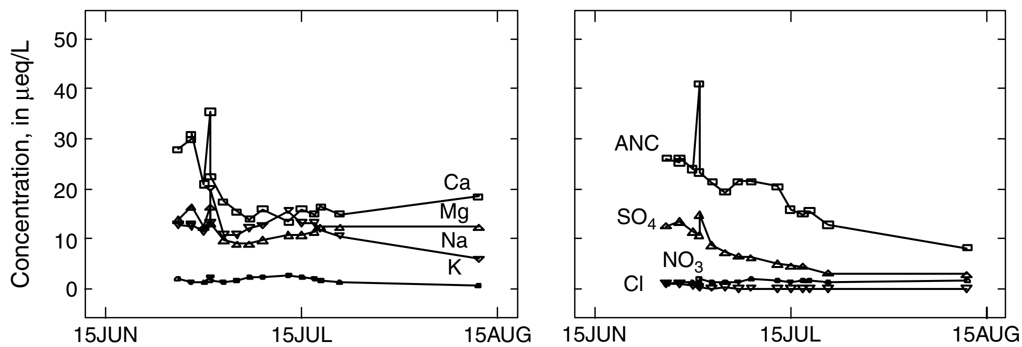

Figure 6. Concentrations of Ca, Mg, Na, K, ANC, SO_4, NO_3 and Cl in Pond 98-18 during 1999

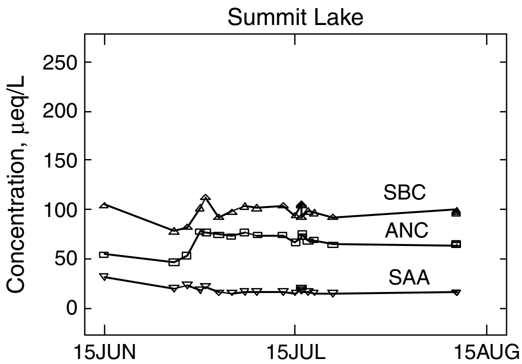

Figure 7. Sum of base cations (SBC), sum of acid anions (SAA) and acid-neutralizing capacity (ANC) for Summit Lake, Buffalo Pass, during 1999. Units are μeq L^{-1}

followed a similar seasonal pattern, with a range of 79–113 μeq L^{-1}. Sum of acid anions was 31 μeq L^{-1} on 15 June then decreased and remained at 15–20 μeq L^{-1} throughout most of the summer.

Dissolved organic carbon

Dissolved organic carbon (DOC) was measured in ponds and lakes at Buffalo Pass during summer months of 2000–2002. The DOC values for the ponds ranged from 0·6 to 34·7 mg L^{-1} with a median of 7·7 mg L^{-1} ($n = 44$); for the lakes, values ranged from 0·9 to 9·4 mg L^{-1} with a median of 3·3 ($n = 97$).

DISCUSSION

Sensitivity to acidification

At a coarse scale, sensitivity to acidification typically has been classified on the basis of bedrock geology because the reactivity of minerals in the geological parent material determines the supply of weathering products (e.g. Omernik and Powers, 1983). The MZWA and adjacent areas are underlain by granitic and volcanic bedrock types that are resistant to weathering and produce little ANC (Snyder, 1980a,b; Turk and Campbell, 1987). At a finer scale, supply of weathering products also may be affected by heterogeneity of mineralogy within mapped bedrock types, differences in surficial geology within mapped bedrock types, and varying degrees of physical weathering and soil development. An earlier study of lake sensitivity in the MZWA demonstrated the importance of surficial geology: the watersheds of sensitive lakes (low ANC) were dominated by exposed granitic bedrock, whereas the watersheds of less-sensitive lakes (higher ANC) had considerable glacial till containing substantial reservoirs of shallow ground water (Turk and Campbell, 1987). Compared with bedrock, till had a greater supply of weathering products as a result of more surface area for mineral weathering, and till discharged more groundwater to the lakes owing to higher hydraulic conductivity. Basin characteristics such as steepness, vegetative cover and surficial debris have also proven useful to predict ANC and base cations in alpine/subalpine basins of the nearby Front Range (Clow and Sueker, 2000), as these terrain features also play a role in controlling the supply and transport of weathering products in watersheds.

On basalt mesas of the Flattops Wilderness Area and Grand Mesa in Colorado, where geology is relatively uniform, altitude and topographic position have been used to predict lake ANC (Turk and Campbell, 1984). The concept of hydrological landscapes has since been developed to characterize the physical interaction of groundwater and surface water in systems ranging in area from square metres to thousands of square kilometres (Winter, 2001). The physical hydrology determines the hydrological flowpaths that control transport of weathering products in groundwater to surface waters. A perched pond on top of a knoll or ridge should receive little input of groundwater (and associated weathering products), whereas a pond in a topographic low area or at the toe of a hillslope should receive greater inputs of ground water and have higher concentrations of weathering products to buffer acidic deposition (Figure 8).

Sum of base cations and ANC varied greatly among ponds in this study, even for ponds located within tens of metres from one another and having watersheds with similar geological parent material, soils and vegetation. Some of this may be explained by small unmapped variations in geology, but the variability in ANC of adjacent ponds indicated that position in the hydrological landscape was controlling inputs of soil water and groundwater, which are the primary source of base cations and ANC. Topographic maps were useful for identification of potentially sensitive features. The most sensitive lake sampled in this study was Summit Lake, which straddles the Continental Divide at Buffalo Pass. At both the Dumont and Buffalo Pass sites, ponds located in lowland areas receive groundwater inputs and had higher ANC than ponds located in upland areas. The ponds near Mount Ethel were identified on topographic maps as upland ponds because of their ridgetop location, and results indicate they are the most sensitive group of ponds examined in this study.

Substantial variability remained in ANC of ponds located within sites that appeared relatively similar on the topographic maps. Among the MZWA ponds, Pond 98-13 and Pond 98-18 are located approximately 300 m from one another, and differences in their landscape position are not apparent on topographic maps. However, field observations indicated potential for groundwater discharge from a hillslope on the north side of Pond 98-13, whereas local relief around Pond 98-18 indicated little potential for inputs of groundwater. Pond 98-13 had ANC that was approximately five times greater than that of Pond 98-18. Because differences of only a few metres in elevation were significant in determining hydrological flowpaths and resulting surface-water chemical composition for individual ponds, quantification of the effect of hydrological landscape position on water chemistry would require an intensive groundwater monitoring network and/or a very high-resolution digital elevation model. Neither of these were practical for this study, nor would they be practical in most studies, especially in remote wilderness locations. However, incorporation of remotely sensed, very high-resolution topographic data in geographical information systems may allow better quantification of hydrological landscape position in future studies.

The Clean Air Act Amendments require resource managers to protect air-quality-related values in Class 1 areas from any impacts of air pollution, which includes protecting all aquatic ecosystems from impacts of acid deposition. In order to protect natural resources without placing unreasonable restrictions on anthropogenic activities that produce air emissions, information is needed to evaluate the current acidification status of surface water and the potential for future impacts from atmospheric deposition in Class 1 areas. The results of this study indicate that acidification models based on stream and lake data are inadequate to predict the sensitivity of small lakes and ponds to deposition, because the quality and resolution of spatial data sets is inadequate and the processes controlling chemistry of ponds are different from those controlling streams and lakes. Monitoring of these systems is needed, however, it is usually impractical to sample all of the ponds and lakes in an area, so it is useful to be able to identify the most sensitive landscapes within an area and

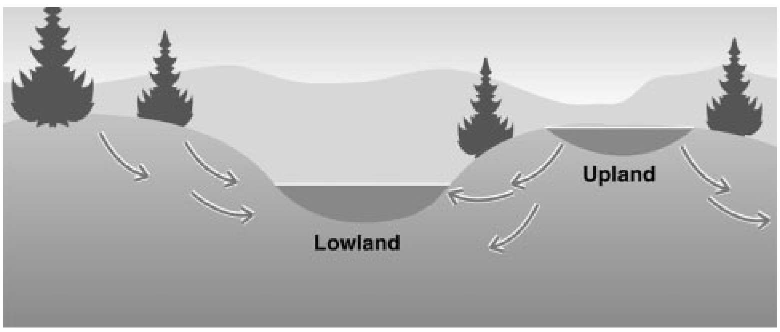

Figure 8. Schematic diagram showing the role of hydrological landscapes in controlling groundwater effects on hydrology and chemistry of ponds

develop a stratified sampling design that focuses on the most sensitive resources. The qualitative approach described, combining use of existing topographic and geological maps with field observation of hydrological landscapes, should be useful for designing cost-effective inventories of aquatic resources in areas where little information is available regarding response of aquatic ecosystems to acidic deposition.

Effects of episodic acidification

The predicted meltwater pH values were quite acidic (Figure 2), and if they were not neutralized by weathering products, they would be expected to cause adverse biological effects in aquatic species. None of the ponds were so acidic during the open-water season, indicating that the acid in meltwater was neutralized by contact with soil or groundwater or by biogeochemical processes in the ponds themselves (Figures 4, 5 and 7). The most intensive measurements were performed during 1999, when the acidity of the snowpack (expressed as concentration of hydrogen ions) was about one-half of that measured in 1998. The reason for the less-acidic snow in 1999 is uncertain, although emissions may have been reduced at one of the local power plants by installation of emissions controls. Dilution of weathering products was the predominant mechanism producing low ANC during spring. An acidic pulse from snowmelt was not evident in surface-water chemistry during the open-water season. A pulse may have occurred prior to the ponds becoming ice-free, it may have been 'smeared' by spatial variability in the timing of snowmelt, or acids may have been neutralized before reaching surface water.

Kiesecker (1991) reported pH values as low as 4·82 in the Dumont ponds during his studies in 1989–91, although ANC was also measured on a subset of the ponds in 1990 and the lowest reported value was 140 μeq L^{-1}. The lowest values measured in this study at the Dumont ponds were 5·68 units for pH and 22 μeq L^{-1} for ANC, which did not support low pH conditions such as those reported in the earlier study. Precipitation chemistry measured at Buffalo Pass does not indicate a significant difference in acidity of deposition between the periods (NADP/NTN, 2001), so it appears likely that the low pH values in the earlier study resulted from differences in measurement technique and the difficulty of accurately measuring pH in dilute waters.

At current levels of atmospheric deposition, neither chronic nor episodic acidification is an immediate threat to any of the Dumont ponds or to the Buffalo Pass lakes. Although these features would be considered sensitive to acidification under many classification schemes, they do not appear to be immediately threatened by chronic or episodic acidification because even at minimum ANC values, the ponds are capable of neutralizing acidity equal to twice the concentration of acid anions currently measured in atmospheric deposition.

In contrast, some Buffalo Pass ponds and all Mount Ethel ponds that were measured are potentially threatened by chronic and episodic acidification. In these ponds, SBC and ANC were relatively low, and acids from atmospheric deposition were important in determining ANC and pH. Sum of acid anions was equal to or only slightly less than ANC in the low-ANC ponds, indicating that acidity resulting from anthropogenic emissions probably has resulted in substantial reduction of ANC. Although there was no measurement of ANC equal to or less than zero, there were eight measurements less than 10 μeq L^{-1} and 44 measurements less than 20 μeq L^{-1}. Given the variability of chemical composition of the ponds in space and time, it is possible that some ponds in the MZWA become acidic for brief periods during snowmelt or summer rainstorms. Some high-ANC ponds such as Pond 98-13 and George Lake showed a large increase in concentration of SBC and ANC after snowmelt, probably from input of groundwater rich in weathering products. In contrast, the low-ANC ponds such as Pond 98-14 and Pond 98-18 had relatively constant concentrations of SBC and ANC, indicating that there was little or no input of groundwater. With little input of groundwater or surface water, the chemistry of these ponds probably is dominated by atmospheric deposition and biogeochemical processes that occur in the water column and in bottom sediments.

Removal of acid anions during the period after snowmelt was similar in all the ponds irrespective of their position in the hydrological landscape. If this removal was occurring in the soil- or ground-water system, differences in removal rates would be expected to vary with hydrological landscape position. Instead, this removal probably occurs within the ponds themselves.

Based on mass balance and seasonal patterns in sulphate concentration, it was previously believed that sulphur behaved relatively conservatively in most low-ANC alpine/subalpine lakes of the Rockies (Turk and Spahr, 1991). However, recent studies using sulphur isotopes indicate that biogeochemical cycling of sulphur can be significant (Brock, 1997; Michel *et al*., 2000, 2002). In this study, sulphate was removed from the water column in most ponds as the season progressed. Sulphate reduction that occurs in anoxic pond and lake sediments consumes acidity and may be contributing to ANC during late summer. Sulphate removal was greater in the ponds than in the lakes (Figures 5 and 7) (Turk *et al*., 1993), probably because the shallow ponds have a greater ratio of bottom surface (sediment) area to water volume. Biogeochemical processes at the sediment–water interface are also of greater importance in the ponds because fluxes of surface water and groundwater are small compared with most lakes, creating a longer hydrological residence time for reactions to occur.

Nitrate also was removed during the summer months. Nitrogen is probably assimilated by both rooted vegetation and diatoms, as there was also a decrease in silica concentrations. Assimilation of nitrogen during photosynthesis consumes hydrogen ions and would contribute to higher ANC. Denitrification also may occur in anoxic sediments and would contribute ANC during later summer. Low nitrate concentrations indicated no net nitrification that would cause a decrease in ANC. The nitrogen dynamics in these systems appear to be very different from alpine/subalpine watersheds in the Front Range, where similar N deposition rates have caused nitrogen saturation and substantial export of nitrogen in surface waters (Williams *et al*., 1996). This difference is probably because watersheds of the lakes and ponds in this study are less steep, are more vegetated, have better developed soils, and contain less talus than those in the Front Range (Clow and Sueker, 2000). The long-term effects of elevated N deposition on terrestrial ecosystems in the MZWA are unknown because there is little data on C : N ratios and N cycling processes in soil and vegetation to compare the nutrient status of the MZWA with other areas with documented effects of nitrogen deposition (Baron *et al*., 2001).

Surprisingly, ANC remained low throughout summer in the most sensitive ponds, despite constant or slightly increasing base cations and decreasing acid anions. This resulted in a significant anion deficit in the charge balance of these samples (20–30 μeq L^{-1} for the sensitive ponds). The anion deficits indicate that during summer, organic acids contribute significantly to acid-base chemistry of the ponds, unlike lakes, which are dominated by inorganic ions.

Dissolved organic carbon (DOC) was not measured during the intensive study, however, in measurements in a subset of the lakes and ponds during 2000–2002, there was a strong relationship between anion deficit and DOC, indicating that the anion deficit represents dissolved organic acids. Similar results were found in a study of three vernal ponds in the Front Range of Colorado, where DOC ranged between 5 and 20 mg L^{-1}, and the contribution of DOC to acid-base chemistry was significant (Vertucci and Corn, 1996). The regression slope of anion deficit on DOC for the MZWA lakes was about twice that for the ponds (Figure 9), indicating that the source and nature of organic matter was different in the ponds compared with the lakes. Distinct sources of carbon were also supported by measurements of ultraviolet (UV, 254 nm) light absorbance on samples collected in 2002: The slope of the relation between UV absorbance and DOC was 0·032 cm^{-1} in 33 lake samples, compared with 0·012 cm^{-1} in 12 pond samples ($r^2 > 0{\cdot}85$; $p < 0{\cdot}001$ for each relationship). The UV absorbance data indicate that low to moderate organic matter concentrations in the lakes contained more aromatic acids probably derived from soils (allochthonous, or terrestrial sources), whereas higher concentrations in the ponds contained more aliphatic organic acids derived from primary productivity (autochthonous, or aquatic sources)(McKnight *et al*., 1994; Aiken and Cotsaris, 1995). The difference in sources of organic matter in lakes versus ponds reflects a fundamental difference in hydrology and biogeochemical cycling between the two types of systems.

CONCLUSIONS

Numerous ephemeral ponds in the Mount Zirkel Wilderness Area are extremely sensitive to acidic deposition. Many of the ponds are much more sensitive than lakes that have been described previously (Turk and Campbell,

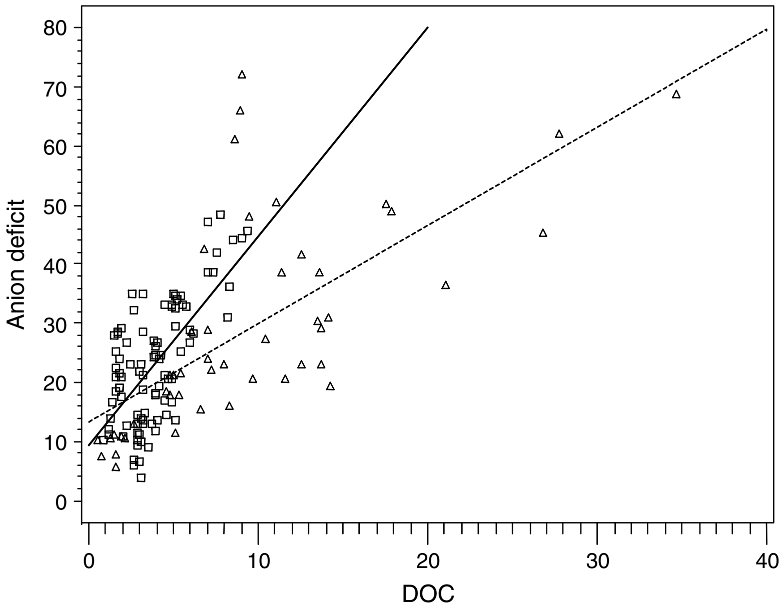

Figure 9. Relation between anion deficit (μeq L^{-1}) and dissolved organic carbon (DOC, in mg L^{-1}) in samples collected during 2002. Lakes are represented by squares and solid regression line (Lake anion deficit = 9.3 + 3.5 (DOC), $r^2 = 0.45$, $p < 0.0001$); ponds are represented by triangles and dashed regression line (Pond anion deficit = 13.3 + 1.7 (DOC), $r^2 = 0.46$, $p < 0.0001$)

1987) and in this study. Although no ponds or lakes had net acidity during the study, a number of the ponds had ANC of 3–20 μeq L^{-1}, making these aquatic ecosystems among the most sensitive to acidification in the World. In Europe, critical loads of sulphur and nitrogen deposition are often defined as amounts that cause ANC to drop below 20 μeq L^{-1} (Curtis *et al*., 2002). The current combination of natural organic acidity and atmospheric deposition indicate that many of the ponds have little capacity for assimilation of additional acids, and that these systems should be considered when assessing ecosystem impacts of acidifying deposition.

About 15% of all pH values measured in this study were less than 6, which indicates a range where adverse impacts to aquatic invertebrates, fish, or amphibians are possible (Harte and Hoffman, 1989, Baker and Christensen, 1991; Kiesecker, 1991, 1996). Most of the low pH values were measured in the Buffalo Pass and Mount Ethel ponds, within the MZWA. In the experiments done in the companion study, relationships between ANC and hatching success for salamanders and chorus frogs at both sites were confounded by differences in water temperature between the ponds, so the higher mortality rate of salamanders in the low-ANC ponds at Buffalo Pass was not statistically significant; however, sublethal effects of acidity on amphibian populations were not examined (Muths *et al*., 2003).

Emissions controls that have been installed or proposed for local coal-fired power plants may reduce atmospheric deposition of sulphuric and nitric acid in the MZWA. Acid-neutralizing capacity during snowmelt was approximately equal to SAA in the sensitive ponds; therefore, any substantial changes in atmospheric deposition of HNO_3 and H_2SO_4 should result in a measurable change in pond chemistry. A decrease in H_2SO_4 deposition would result in higher ANC during snowmelt; however, the effects on ANC later in the summer are uncertain because ANC could decrease if SO_4 reduction is currently contributing ANC. A decrease in HNO_3 deposition also would result in higher ANC during snowmelt but probably would have little effect on acidification status later in the summer. A change in deposition of either ammonia or nitrate also could affect primary productivity and in turn affect other biological processes. It is unknown what nutrients limit productivity of ponds and lakes in the MZWA, other high-elevation aquatic ecosystems in the western USA are limited by nitrogen, phosphorous, or both (Fenn *et al*., 2003). The MZWA provides a unique opportunity

Table I. Sensitivity to acidification. Station identity includes location in NAD83 datum, formatted as latitude and longitude in degrees, minutes, seconds (ddmmssdd-mmss00)

Water body group	Station identity	Station name	Acid-neutralizing capacity (μeq/L)			Number of samples
			Minimum	Mean	Maximum	
Buffalo Pass ponds	402738106393500	Dos Equis Pothole (XX)	12	13	15	2
	403254106403400	Pond 98-05	11	24	58	7
	403255106405200	Pond 98-08	40	47	56	3
	403306106411400	Pond 98-07	11	20	30	16
	403308106402100	Pothole B-14	24	35	43	8
	403310106410400	Pond 98-06	40	49	68	6
	403313106411100	George Lake	53	89	116	21
	403322106410500	Pond 97-02	96	142	210	4
	403325106410500	Pond 97-5	113	175	242	3
	403325106411100	Pond 98-10	48	54	59	3
	403345106402900	Pond 98-01	20	29	36	3
	403350106402900	Pond 98-02	41	49	56	3
	403400106414700	Pond 98-17	47	104	138	5
	403404106403700	Pond 98-09	115	167	220	2
	403408106420100	Pond 97-07	17	35	49	5
	403409106420800	Pond 97-06	64	76	105	6
	403412106402800	Pond 98-03	23	29	36	3
	403414106420900	Pond 97-08	107	114	122	2
	403418106403900	Pond 98-04	28	41	71	4
	403424106413300	Pond 97-04	54	72	82	5
	403427106412700	Pond 98-16	46	65	77	6
	403435106411800	Pond 98-18	3	19	41	20
	403437106411600	Pond 98-14	7	15	26	20
	403437106411800	Pond 98-15	34	51	82	19
	403440106411100	Pond 98-11	23	165	279	6
	403441106410200	Pond 98-19	37	47	58	2
	403441106410400	Pond 98-12	43	59	73	5
	403442106411500	Pond 98-13	55	165	247	20
	403500106415100	Pothole B-5	33	45	52	4
	403520106424100	Pothole B7	19	21	23	2
	403522106424300	Pothole B-6	23	23	23	1
Dumont ponds	402350106373700	Dumont Pond 1	150	150	150	1
	402353106373100	Dumont Pond 2	164	164	164	1
	402358106374600	Dumont Pond 7	76	118	156	7
	402359106375000	Dumont Pond 8	86	112	176	5
	402359106380100	Dumont Pond 12	82	105	128	5
	402400106375800	Dumont Pond 11	44	62	86	27
	402401106375400	Dumont Pond 9	99	108	120	3
	40240110 6375900	Dumont Pond 40	46	51	55	2
	402402106375700	Dumont Pond 13	78	95	111	7
	402403106374400	Dumont Pond 6	84	113	138	6
	402403106380100	Dumont Pond 25	44	66	89	19
	402404106374100	Dumont Pond 3	382	389	397	2
	402404106374600	Dumont Pond 5	94	113	147	5
	402404106375400	Dumont Pond 10	76	112	174	25
	402404106380400	Dumont Pond 35	51	64	73	3
	402405106374700	Dumont Pond 4	77	112	128	4
	402405106375000	Dumont Pond 26	59	96	155	22
	402405106375600	Dumont Pond 15	77	119	204	25

(*continued overleaf*)

Table I. (*Continued*)

Water body group	Station identity	Station name	Acid-neutralizing capacity (μeq/L)			Number of samples
			Minimum	Mean	Maximum	
	402405106375800	Dumont Pond 16	66	83	104	4
	402406106375400	Dumont Pond 14	22	45	77	23
	402406106380600	Dumont Pond 24	69	98	114	3
	402406106381000	Dumont Pond 39	57	59	61	2
	402407106374700	Dumont Pond 29	142	168	193	3
	402408106380200	Dumont Pond 36	41	51	58	3
	402409106250000	Dumont Pond 38	45	50	56	3
	402410106380200	Dumont Pond 41	41	41	41	1
	402410106380400	Dumont Pond 23	87	98	109	2
	402410106383900	Dumont Pond 31	52	57	63	4
	402411106383100	Dumont Pond 18	45	62	93	6
	402412106380000	Dumont Pond 50	139	139	139	1
	402412106383600	Dumont Pond 20	40	55	71	5
	402413105380300	Dumont Pond 22	79	79	79	1
	402413106383100	Dumont Pond 19	45	46	47	2
	402413106383500	Dumont Pond 37	34	42	53	3
	402414106380500	Dumont Pond 33	42	57	69	3
	402414106380600	Dumont Pond 34	84	103	119	4
	402414106383400	Dumont Pond 32	28	37	47	5
	402415106380800	Dumont Pond 21	99	157	205	5
	402431106383400	Dumont Pond 17	66	98	144	6
Lakes	402315106350200	Dumont lake	221	262	338	8
	402654106393800	Lost Lake	77	80	83	2
	402715106391300	Fish Hook Lake	238	246	253	2
	402809106393200	Little Lost Lake	109	120	131	2
	402822106394400	Round Lake	234	241	248	2
	402833106412400	Long Lake Res.	102	117	127	4
	403044106415400	Dinosaur Lake	77	82	87	2
	403245106404900	Summit Lake	46	66	78	34
	403306106403800	Jonah Lake	64	93	115	8
	403325106402600	Whale Lake	73	91	107	5
	403330106405100	Martha Lake	73	121	172	7
	403414106403700	Lake Albert	63	76	88	3
	403700106470000	Fish Hawk Lake	60	60	60	1
	403743106455300	Snowstorm Lake	61	61	61	1
	403744106444700	Rosa Lake	53	53	53	1
	403759106445100	Mirror Lake	83	83	83	1
	403759106452300	Margaret Lake	73	73	73	1
Mount Ethel ponds	403720106412000	Pond 99-01	10	10	10	1
	403721106413200	Pond 99-05	13	13	13	1
	403722106412000	Pond 99-02	24	24	24	1
	403722106413100	Pond 99-04	14	14	14	1
	403723106412400	Pond 99-03	17	17	17	1
	403724106413900	Pond 99-07	33	33	33	1
	403725106413400	Pond 99-06	17	17	17	1
	403729106414200	Pond 99-08	34	34	34	1

to test source–receptor relationships as local power plant emissions continue to decrease in coming years. Decreased acidifying emissions from local power plants may help reduce acidic deposition and increase ANC and pH in surface waters in the MZWA. However, this improvement could be offset by other changes in emissions resulting from regional population growth, agricultural activities and energy development.

In some respects, the ponds and lakes in this study represent a worst-case scenario for episodic acidification in the Rocky Mountain region, because of the combination of the most acidic precipitation in western North America and the sensitivity of the watersheds to acidification. However, this hypothesis has not been tested by examining ponds in other parts of the region that also could be sensitive to acidification. In particular, additional sources of acidity, such as organic acid production and weathering of natural exposures of pyritic minerals, could result in more acidic conditions with similar or less acidic atmospheric deposition. Differences in assimilation of nitrogen and sulphur compounds in terrestrial and aquatic ecosystems also are important. Biogeochemical cycling in the water column and at the sediment–water interface are poorly understood and confound prediction of the chemical and biological response of ponds to acidic deposition as well as other stressors such as extremes of climate.

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