

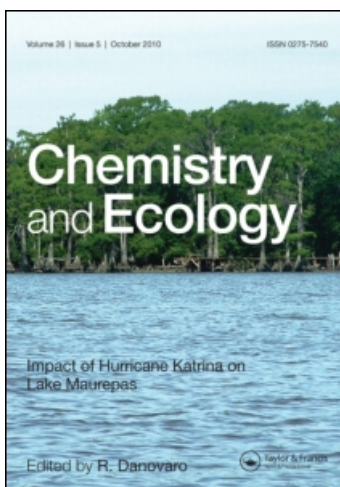
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## RESEARCH ARTICLE

# Acidity of machine-made snow and its effect on pH and aluminum speciation in New England streams during spring thaw

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The pH of machine-made snow and its effect on an acid-sensitive watershed in Vermont were studied. Spring runoff from snowmaking was found to be less acidic and to contain less dissolved inorganic aluminum. Dissolved inorganic aluminum has been associated with damage to aquatic life. The extensive use of machine-made snow by the ski industry in most of the northeast region of the US may be beneficial to aquatic life.

**Keywords:** acidity; aluminum; machine-made snow; pH; snowmelt; speciation

### 1. Introduction

The ski industry in New England is heavily dependent on machine-made snow [1]. Interestingly, the northeast US is an area also noted for the problems of acid precipitation. Numerous studies including [2–7] have noted a pulse of low pH runoff during spring snowmelt. These studies demonstrate that the chemistry of spring snowmelt is quite complex. However, all studies agree that the acid pulse is also associated with a high concentration of free inorganic aluminum in stream water during the spring snowmelt. This form of aluminum is particularly toxic for many aquatic species, especially trout [2]. Details of aluminum toxicity are complex and depend on the form of aluminum present, the pH, and the life stage of the organism [8,9,19]. In general, any activity which reduces the acidity would have a potential benefit for aquatic species. The goals of this study were to compare the acidity of machine-made snow to natural snow and to study its effects on aluminum speciation in the spring runoff.

Machine-made snow is normally made using surface and ground water. At most ski areas this is taken directly from streams or ponds. In all cases, the water used for making snow is exposed

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to soil and sediment and is therefore buffered with dissolved minerals and typically has a pH in the range of 6–8. Environmental pollutants, due primarily to emissions from coal burning power plants in the mid-western states, make the precipitation in much of the northeast US considerably more acidic than natural precipitation. The average pH of precipitation in Vermont is slightly less than 4.4 [10]. The hypothesis of this study was that machine-made snow would be less acidic than natural snow and thus the spring snowmelt from machine-made snow would contain lower levels of toxic inorganic aluminum.

An estimation of the probable change in acidity was made based on the sum of the total amount of water used in snowmaking in the study site (1.9 billion litres) [11] plus the amount of water from natural precipitation (5.4 billion litres) obtained from the total melted precipitation (220 cm) [12] multiplied by the total snowmaking area (2.45 km<sup>2</sup>) [11]. The acidity of natural snow was based on a pH of 4.4 and the acidity of the snowmaking water was based on a pH of 7.0. Our estimation indicated that when using machine-made snow there would be about 40% less acid present in the spring runoff.

The pH, along with the presence of natural ligands has a major impact on the speciation of aluminum in natural water. Of the various forms, free inorganic aluminum has been shown to be most toxic to fish and is the form that predominates at higher acidity (lower pH values) [2]. Aluminum in colloidal particles and that bound to organic ligands and fluoride is less toxic. Free inorganic aluminum, complexed dissolved aluminum, and total aluminum were analysed in our study using frequently cited literature methods [13].

## 2. Experimental

### 2.1. The site

Two ski areas in south-central Vermont which use extensive snowmaking were selected for this study. Okemo Mountain (N 43.4, W 72.7) was the primary study site with additional samples obtained from Killington Mountain (N 43.6, W 72.8). Sites for snow collection were selected both in the ski area and on another part of the mountain several miles away from any snowmaking activity. Sites were selected to pair natural and machine-made snow at several altitudes from 360 to 920 m. Machine-made snow was collected in the early winter months prior to any significant natural snow. Some of the machine-made snow samples also contained a small amount of natural snow.

Six streams were identified on Okemo Mountain, three draining the ski area and three which involved no machine-made snow. All six streams were monitored for the year of the study. These six streams varied considerably in watershed, flow, and acidity. Based on this analysis, two first order streams, Giant Brook (N 43.365, W 72.739, 550 m a.s.l.) and Coleman Brook (N 43.422, W 72.719, 366 m a.s.l.) were chosen as the primary study sites, and samples were obtained upstream from most human activity other than snowmaking. The two streams chosen were both highly acidic and similar as detailed below. Giant Brook is far removed from any snowmaking while Coleman Brook drains a significant portion of the Okemo Mountain Resort ski area. Discharge was monitored using Global Water Discharge Monitors from March through May and stream depth profiles were obtained.

Both flow into the Black River in Ludlow VT. The watersheds of these two streams were estimated using ArcMap 9.1 from 50 meter contour GIS layers [14]. Both watersheds are roughly 150 to 180 hectares in area. The fraction of the Coleman Brook watershed which is covered by snowmaking was estimated from aerial photos as approximately 20% of the total area. Total precipitation from 1 December 2004 to 7 May 2005 was reported at the Ludlow weather station [11] as 220 cm.

## 2.2. Soil and bedrock in the watershed of the primary study streams

The tree cover at the two sites was evaluated using a colour orthophoto [15]. Both sites contain about equal coverage of conifers and hardwoods. The Giant Brook watershed had approximately 5% open land as evaluated from the aerial photo. The presence of ski trails on the Coleman Brook site was estimated to cover approximately 20% of the watershed. The remainder of the site is forested.

The bedrock geology of the sites was evaluated using GIS layers from the USGS [16–18] and the soil was evaluated using GIS layers from VCGI [13]. Unfortunately, the GIS data available was not complete and quantitative data on relative areas of each soil and bedrock type could not be obtained. Bedrock geology for all sites consisted of metamorphic schist and gneiss with little marble. No significant differences in bedrock were observed. Both Giant Brook and Coleman Brook include outcrops of schist and gneiss in their stream beds. A detailed list of bedrock at each site is provided in Table 1.

Soils were also found to be generally similar at the two primary sites. Detailed soil descriptions were examined using information from the Natural Resources Conservation Service [19] and a complete list of soil types is included in Table 2. Most soils in both watersheds were glacial and well drained. All of the soils in the Giant Brook watershed are very well drained and range from strongly to extremely acidic.

The general conclusion is that the vast majority of all the soils on both sites are acidic and that there is no obvious reason to expect one watershed to be significantly more acidic than the other.

## 2.3. Sampling methodology

Snow samples were obtained using a 12 inch AMS Soil recovery probe (Forestry Supply Inc) with butyrate plastic liners. When snow depth was less than 12 inches, multiple cores were taken and combined in one tube. Plastic tubes were capped with polyethylene caps and stored frozen until analysis.

Water samples were grab samples taken using a polyethylene sampling cup. All samples were taken in late afternoon. Samples were initially stored in WhirlPac Bags (Forestry Supply) and refrigerated until analysis, which was done within 24 hours.

Water samples were taken over a full year with a sampling frequency of once a month prior to the beginning of the spring thaw and approximately every 10 days from the beginning of the thaw until there was no snow remaining.

Water for snowmaking at the Okemo site is drawn from a man-made reservoir and from a pond filled from the Black River during high flow periods. Access to these sites during the winter was restricted, however, one sample obtained during the summer was found to have a pH of 6.54,

Table 1. Bedrock in the watershed of the study streams.

Coleman Br.	Giant Br.	LITH	Description
*	*	Ya	Amphibolite
*		Yap	White feldspar-rich aplite
*	*	Ybg	Well-layered, biotite-quartz-plagioclase gneiss and amphibolitic gneiss
*	*	Ycs	Calc-silicate gneiss, minor marble, diopside-hornblende rock, actinolite marble
*	*	Yp	Pegmatite
*	*	Yq	Thin-bedded, white to grey, vitreous quartzite and garnet quartzite
*	*	Yrs	Rusty quartz-muscovite $\pm$ chlorite schist to richly garnetiferous quartz schist
*		Yta	Tourmaline aplite
	*	Ylq	Massive, vitreous, well-jointed quartzite on Ludlow Mountain
	*	Yt	Biotite trondhjemite gneiss at Terrible Mountain

Table 2. Soil types in the watershed of the study streams.

Coleman Br.	Giant Br.	MUSYM	MUNAME
	*	11D	Marlow fine sandy loam, 15–35% slopes, very stony
	*	11E	Marlow fine sandy loam, 35–60% slopes, very stony
*		122B	Lyme fine sandy loam, 2 to 8 percent slopes, very stony
*		128C	Rawsonville-Houghtonville complex, 8–15% slopes, rocky
*	*	128D	Rawsonville-Houghtonville complex, 15–35% slopes, rocky
*		129F	Killington-Rawsonville complex, 35–70% slopes, very rocky
*		12D	Tunbridge-Lyman complex, 15–35% slopes, very rocky
*		12E	Tunbridge-Lyman complex, 35–60% slopes, very rocky
	*	18C	Peru, Skerry, and Colonel soils, 8–15% slopes, very stony
	*	18D	Peru, Skerry, and Colonel soils, 15–35% slopes, very stony
*		29A	Grange very fine sandy loam, 0–3% slopes
*	*	31B	Cabot loam, 0–8% slopes, very stony
*		31C	Cabot loam, 8–15% slopes, very stony
*		58D	Berkshire-Tunbridge complex, 15–35% slopes, very stony
*		58E	Berkshire-Tunbridge complex, 35–50% slopes, very stony
*	*	59C	Rawsonville-Houghtonville complex, 8–15% slopes, rocky
*	*	59D	Rawsonville-Houghtonville complex, 15–35% slopes, rocky
*		59E	Rawsonville-Houghtonville complex, 35–60% slopes, rocky
*	*	60D	Glebe-Stratton complex, 15–35% slopes, very stony
*	*	60F	Glebe-Stratton complex, 35–70% slopes, very stony
*		61D	Ricker-Londonderry-Stratton complex, 15–35% slopes, very rocky
*		61F	Ricker-Londonderry-Stratton complex, 35–70% slopes, very rocky
*		62D	Hogback-Rawsonville complex, 15–35% slopes, very rocky
*		63C	Berkshire and Monadnock fine sandy loams, 8–15% slopes, very stony
*		64C	Colton fine sandy loam, 8–15% slopes
	*	74D	Mundal fine sandy loam, 15–35% slopes, very stony
*	*	132C	Glebe-Stratton complex, 8–25% slopes, very stony
*	*	132E	Glebe-Stratton complex, 25–60% slopes, very stony
*		134F	Stratton-Londonderry-Ricker complex, 15–80% slopes, very rocky
*		150A	Peacham muck, 0–8% slopes

close to 7 as expected. The spring snowmelt included four major peaks in the discharge profile. These occurred on 2 April at 23:00 EST, 8 April at 0:00 EST, 24 April at 2:00 EST, and 28 April at 3:00 EST. Samples were obtained on the falling limb of the first two events and on the rising limb of the third. The fourth event was not sampled. Details on precipitation and discharge are shown in Table 3.

#### 2.4. Statistical analysis

Statistical analysis was performed using Microsoft Excel 2003. Two sample t-tests were used assuming either equal or unequal variances depending on the observed variance of the data. Paired two-sample t-test for means was used to compare stream data paired by date of sample collection. The error bars shown in the stream data figures for Giant and Coleman Brooks are based on the long term coefficient of variation (CV) of the standard solutions over the course of the study and are thus a conservative estimate of the errors for individual measurements.

#### 2.5. Analytical methods

The analysis of aluminum was performed using the Flow Injection Analysis (FIA) pyrocatechol violet (PCV) method which was modified for use on discrete samples [20]. All reagent solutions were prepared in 18.3 mOhm DI water (Barnsted). To 2.0 ml aliquots of water samples were added in order 64  $\mu$ l phenanthroline stock solution (30 g hydroxylamine HCl and 0.30 g phenanthroline

Table 3. Precipitation and hydrologic conditions at sample points for stream study.

Date	Precipitation	Stream flow
18 Sep 2004	1.02 cm 17 Sep	Falling limb
	3.56 cm 18 Sep	
13 Nov 2004	None	Base flow
17 Feb 2005	None	Base flow
22 Mar 2005	None	Base flow
4 Apr 2005	Spring melt	Falling limb after highest discharge for spring season
	4.83 cm 28 Mar	
	2.54 cm 2 Apr	
8 Apr 2005	None	Falling limb second major thaw
23 Apr 2005	3.81 cm 23 Apr	Rising limb third major thaw
	2.54 cm 24 Apr	
4 May 2005	None	Base flow
13 May 2005	None	
23 Oct 2005	1.52 cm 22 Oct	Falling limb
	1.02 cm 23 Oct	

\*National Climate Center Data, 2006.

in 100 ml), 120  $\mu$ l PCV (0.1875 g PCV in 100 ml) and 1.60 ml buffer (20 g hexamethylenetetramine in 100 ml) in a 1.0 cm path length cell suitable for UV analysis. The cell was capped and thoroughly mixed and the absorbance measured using Ocean Optics USB2000-UV-Vis spectrometer (OOChem software). The aluminum PCV method was calibrated daily using deionised water blank and standard aluminum sulfate solutions ranging from 1.2 to 26  $\mu$ M aluminum. The CV of the absorbance of the 1.2  $\mu$ M standard over the period March to May, 2005 was 10% ( $n = 5$ ) while the CV at 13  $\mu$ M was 5% ( $n = 5$ ).

An IQ Scientific Instruments Model 150 portable pH meter was used for all potentiometric measurements. The pH was measured with an ISFET probe (IQ Scientific Instruments) after addition of 1 M KCl solution at 1% of the sample volume. A series of samples were also measured using a Ross pH electrode (Orion) normally used for acid precipitation studies and the results for the two electrodes were found to agree within 0.02 pH units.

Solid Phase Extraction (SPE) cartridges (Alltech Associates, 200 mg IC-SCX cation exchange) were used in the fractionation procedure to separate anionic complexed forms of aluminum from uncomplexed forms. This replaced the use of ion exchange cartridges in previous studies. All SPE cartridges were conditioned prior to use [21]. The pH of the conditioning buffer was adjusted to pH 6.3 to approximate the average pH of the water samples.

### 3. Results and discussion

#### 3.1. Snow results

In this study the acidity of the various types of snow samples was compared. It appears that there is a 10 fold difference (using unequal variances,  $p = 0.0007$ ,  $n = 11$  machine,  $n = 7$  natural) between the machine-made snow and the natural snow. The average hydrogen ion concentration of natural snow was 15  $\mu$ M (SD 6  $\mu$ M,  $n = 7$ ) and that of machine-made snow was 1.5  $\mu$ M (SD 3  $\mu$ M,  $n = 11$ ). The finding that machine-made snow is an order of magnitude less acidic supports the initial hypothesis of this study. It should be noted that the machine-made snow samples contained some natural precipitation and therefore we would expect that pure machine-made snow would be even less acidic. The machine-made snow varied widely from a high of 8.5  $\mu$ M to a low of 0.3 nM (pH range 5.07 to 9.51) reflecting the presence of some natural snow and variations in snowmaking water.

### 3.2. Stream chemistry

Streams samples were analysed during the spring thaw to determine if the acid pulse could be detected and what the effect would be on inorganic aluminum. Weather and hydrological conditions during sampling are shown in Table 3.

Figure 1 shows results of stream acidity for Giant Brook (natural snow) and Coleman Brook (mix of natural and machine-made snow). Both of these streams were naturally acidic. The acid pulse was clearly observed in both streams during snowmelt in April. The pH values for the average hydrogen ion concentrations of the two streams over the full year of the study were 5.88 and 6.06 (paired t-test shows the difference is not statistically significant,  $p = 0.2$ ,  $n = 10$ ). In contrast, it was found that the acidity of Coleman Brook (mix of machine-made and natural) was significantly less ( $p = 0.02$ ,  $n = 8$ ) than that of Giant Brook (natural snow) during the winter and spring. It should be noted that the October 2004 samples showed a distinct acid pulse. This is entirely consistent with the observation of published studies that the autumn rains produce an acid pulse, particularly after a dry fall [2,22]. The fact that the autumn acidity of Coleman Brook is greater than that of Giant Brook adds support to the conclusion that the higher pH of Coleman Brook during the spring thaw was in fact a real effect of the machine-made snow and not due to soil or bedrock differences between the two sites. The similarity of the pH in the two streams under base flow conditions further supports this conclusion.

Since the decrease in acidity would affect the species of aluminum present, a speciation study of aluminum was conducted as described in the experimental section. The results are shown in Figures 2–4. Figure 2 shows the total aluminum present in unfiltered water samples. The spike in total aluminum during snowmelt in Coleman Brook (machine-made snow) and Giant Brook (natural) is quite clear and appears to be the direct result of the spike in acidity.

The dissolved aluminum shown in Figure 3 is measured after removal of all particulates larger than 0.45 microns. The comparison of dissolved and total aluminum indicates that most of the aluminum was present in a soluble form.

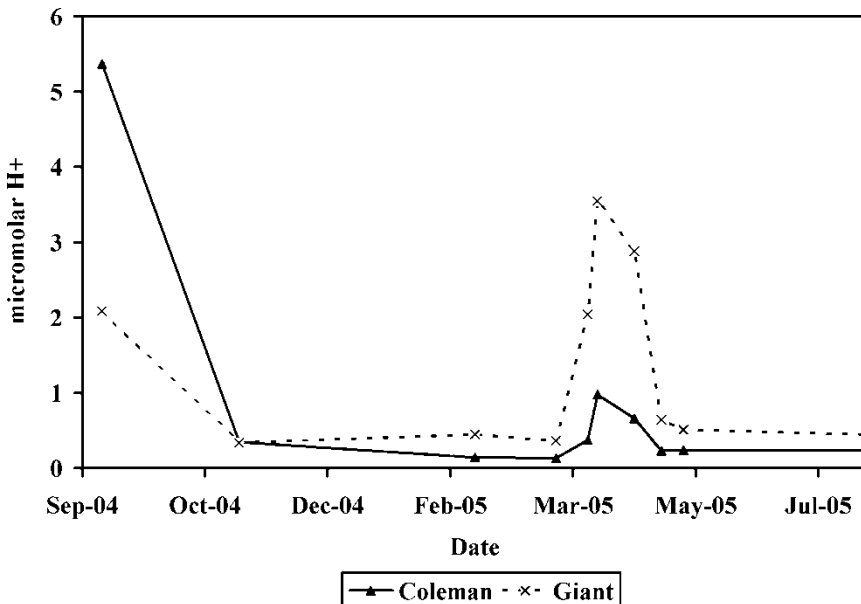


Figure 1. Stream acidity as micromolar hydrogen ion concentration during the 2004–2005 season.

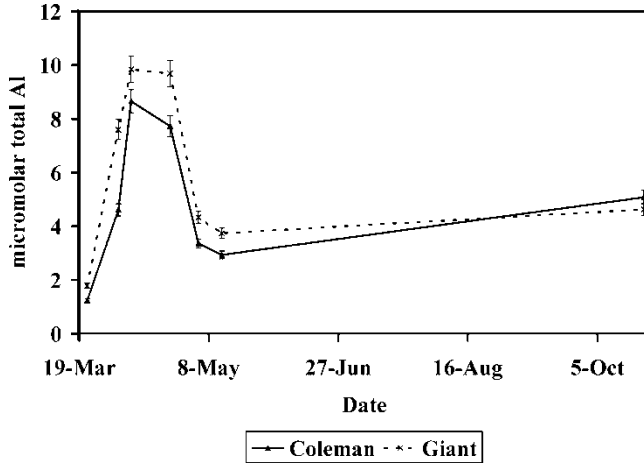


Figure 2. Total aluminum concentrations for streams during the 2005 season.

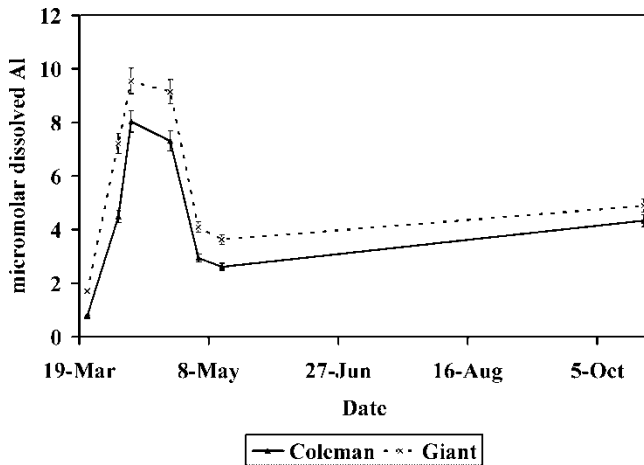


Figure 3. Dissolved aluminum species for streams during the 2005 season.

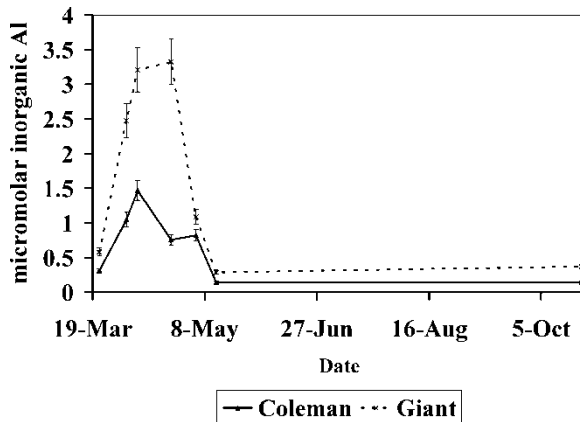


Figure 4. Concentration of free inorganic aluminum ion for streams during the 2005 season.

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Figure 4 shows the free inorganic aluminum in the two study streams. This is aluminum which is not bound to fluoride or to organic ligands (dissolved organic carbon). The inorganic aluminum concentrations for Giant Brook (natural) are almost twice as high as those for Coleman Brook (machine-made). A paired t-test of the two data sets showed that the differences were significant ( $p = 0.02$ ,  $n = 7$ ).

Aluminum data from numerous studies from New Hampshire, New York, and Europe have been reported and summarised [23]. Most of the studies involved streams that were an order of magnitude more acidic than those in this study and as a result, the aluminum concentrations reported were much higher than those reported in this study. The study of a stream in Québec where the observed acid pulse reduced the stream pH from 6.0 to 5.5 [3] is similar to the results observed in Giant Brook. At pH 5.5, the highest inorganic aluminum reported is slightly less than  $2 \mu\text{M}$  [21]. At a pH of 5.5 observed during the acid pulse of Giant Brook, the inorganic aluminum was found to be  $3 \mu\text{M}$ . For Coleman Brook, which reached a minimum pH of about 6, the inorganic aluminum peaked at  $1.5 \mu\text{M}$ , which is very close to values reported in the Québec stream study [3]. The similarity of literature values for inorganic aluminum in streams of pH similar to those of Giant Brook and Coleman Brook supports the accuracy of the present measurements.

#### 4. Conclusions

In conclusion, the results support the initial hypothesis and the model used to predict the impact of snowmaking in this watershed. The data presented in this study suggest that the decrease in acidity observed during the spring thaw containing run-off from machine-made snow is real and is due to the impact of recycled surface and ground water present in machine-made snow but not in natural snow. The decrease of toxic inorganic aluminum observed during the spring thaw is a direct result of this decrease in acidity.

While this data covers only one seasonal thaw, the data are consistent with the known chemistry of aluminum in surface and ground water and the mechanics of snowmaking. The study did not measure alkalinity or dissolved organic carbon. Further study including these additional analytes would be necessary for definitive conclusions and for a complete model of the water chemistry of aluminum in these streams. It would also be helpful to study the chemistry of the water used for snowmaking. This study did not attempt to do this because it would have required cooperation and oversight by the ski areas and could limit scientific independence. The finding in this study suggests that the addition of machine-made snow to natural snow may have a positive impact on fish populations in streams fed from ski areas. This study should encourage ski areas to cooperate in more detailed analysis.

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