

Chemical and isotopic evolution of a layered eastern U.S. snowpack and its relation to stream-water composition

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Abstract The chemical, isotopic, and morphologic evolution of a layered snowpack was investigated during the winter of 1993-94 at Sleepers River Research Watershed in Danville, Vermont. The snowpack was monitored at two small basins: a forested basin at 525 m elevation, and an agricultural basin at 292 m elevation. At each site, the snowpack morphology was characterized and individual layers were sampled seven times during the season. Nitrate and $\delta^{18}\text{O}$ profiles in the snowpack remained relatively stable until peak accumulation in mid-March, except near the snow surface, where rain-on-snow events caused water and nitrate movement down to impeding ice layers. Subsequently, water and nitrate moved more readily through the ripening snowpack. As the snowpack evolved, combined processes of preferential ion elution, isotopic fractionation, and infiltration of isotopically heavy rainfall caused the pack to become depleted in solutes and isotopically enriched. The release of nitrate and isotopically depleted water was reflected in patterns of nitrate concentrations and $\delta^{18}\text{O}$ of meltwater and stream water. Results supported data from the previous year which suggested that streamflow in the forested basin during snowmelt was dominated by groundwater discharge.

INTRODUCTION

In the northeastern United States, snowpack development and evolution encompasses a complex set of processes. Despite frequent very cold weather, air temperatures generally rise above the freezing point several times during the winter. In areas that sustain a snowpack throughout the winter, the resulting freeze/thaw cycles quickly alter the surficial snow layer. Meltwater infiltrates the surface snow and refreezes at depth. Ice layers may also form directly on the snowpack surface when a cold period follows a thaw, or when freezing rain or rain-on-snow occurs. Preferential pathways develop for subsequent movement of water through the pack, controlled in large part by the ice

layers which have been shown to persist through much of the snow melt season in the eastern United States (Albert & Hardy, 1993).

The morphologic evolution of the snowpack affects the chemical and isotopic evolution. Solutes that enter the snowpack in precipitation elute preferentially with the initial meltwater, and when meltwater refreezes within the pack, the solutes are immobilized as well. Thus, solutes tend to move through the pack in stages with successive meltwater pulses (Bales *et al.*, 1993). Initial meltwaters tend to be isotopically lighter than the bulk snowpack (Maulé & Stein, 1990). During melt, the residual snowpack becomes increasingly enriched in ^{18}O because the ^{16}O is preferentially removed in the melt; the equilibrium fractionation between snow and water is 3.5‰ at 0°C (Majoube, 1971). Other processes affecting the $\delta^{18}\text{O}$ of the snowpack include evaporation and condensation of vapor, migration of moisture from underlying soils into the snowpack (Friedman *et al.*, 1991), and additions of late-season isotopically heavy rain. As the pack ripens, interlayer differences in $\delta^{18}\text{O}$ tend to become homogenized (Unnikrishna *et al.*, 1993; Brammer *et al.*, 1995).

The isotopic and chemical behavior of snow has received much attention. Laboratory and field studies have shown that ionic pulses in outflow are due to both grain scale effects: the movement of solutes to the surface of snow grains during metamorphosis (Brimblecombe *et al.*, 1987; Bales *et al.*, 1989; Hewitt *et al.*, 1991) and layering effects (Bales *et al.*, 1989). More pronounced ionic pulses are expected from snow with layers which vary in ionic concentration (Davies *et al.*, 1984). Most studies on snowpack evolution have been done on snowpacks where layering was absent or weakly developed. In this paper, we present field observations of the chemical and isotopic evolution of highly layered snowpacks and melt season effects on streamflow chemistry.

METHODS

This investigation was conducted at two sites about 7 km apart at Sleepers River Research Watershed near Danville in northeastern Vermont. Each site was in a generally south-facing small gauged sub-basin. The W-2 site is an open pasture at 292 m elevation, and the W-9 site is a deciduous forest at 525 m elevation. Each sub-basin is instrumented with a recording stream gauge, four 1-m² snowmelt collectors at various slopes and aspects, and a recording thermistor for air temperature. Wetfall-only precipitation was collected at W-9 at least weekly, and frequently on an event basis. During periods of active melt, the snowmelt collectors were sampled daily and stream water at the sub-basin outlets was sampled at 4-h intervals.

The snowpack was characterized and sampled seven times at each site; both sites were always sampled on the same day. The first four sample visits were made at approximately 3-week intervals leading up to and including peak accumulation in late March. Three additional sampling visits were made approximately weekly during the snowpack ablation period. The snow pit characterization included layer identification, crystal type identification according to ICSI classification (Colbeck *et al.*, 1990), and density, grain size, and snow temperature profiles. Each profile was divided into snow layers not exceeding 18 cm, each with an approximately uniform texture. Each layer was subsampled and composited using a polyethylene scoop; ice layers ≥ 1 cm were included. Thinner ice and crust layers were included with the underlying layer, based

on the assumption that these layers formed within wind crusts originally developed at the surface of freshly fallen snow. An integrated core sample was also collected at each sampling time. All samples were placed in sealable polyethylene bags.

Snow samples were melted at room temperature but not allowed to warm. Snow, snowmelt, and stream samples were kept chilled and were filtered (0.45 μm , cellulose acetate) within 6 weeks of collection. One aliquot was acidified with HNO_3 to pH 2 and analyzed for Ca, Mg, Na, and K by atomic absorption spectrophotometry. An unacidified aliquot was analyzed for Cl, NO_3 , and SO_4 by ion chromatography. To evaluate the chemical evolution of the snowpack, this paper will focus on nitrate. $\delta^{18}\text{O}$ was determined by mass spectrometry. Values are reported in per mil (‰) relative to Standard Mean Ocean Water (SMOW). Analytical precision for $\delta^{18}\text{O}$ is $\pm 0.05\text{‰}$.

RESULTS AND DISCUSSION

Morphologic evolution of the snowpack

The snow season commenced at W-9 in early December. By 21 January, the snow pack was approximately 70 cm deep and consisted of precipitation particles in the top half of the pack, with faceted crystals developing in the lower half (Fig. 1(a)). A thin discontinuous ice layer and a weak basal ice layer in the lower portion of the pack reflected December rain-on-snow events. The early February snow profile showed a distinct ice layer resulting from the 3.2 cm of rain on 28 January. This ice layer was traceable into April (Fig. 1(b)). By peak accumulation, near 18 March, the top portion of the pack consisted of rounded grains and several crust layers, while faceted particles remained in the lower portion of the pack. By April, the dry snow types gave way to rounded clusters and melt-freeze particles. Snow completely disappeared from this site by 25 April.

The open, lower-elevation site, W-2, consistently had less snow than at W-9. Throughout most of the mid-season, a similar layering pattern was evident, but the pack at W-2 became ripe (texturally homogenous) before W-9 (Fig. 1(b)). An ice layer formed at the snow surface after the 28 January rain-on-snow event and was also traceable into April. A basal ice layer was observed in the snowpack during the March measurements. At the time of peak accumulation, the W-2 snow depth was approximately half that of W-9. By 28 March, the faceted particles in the lower half of the pack began to round. In April the snow was composed entirely of wet grains. The snowpack was ripe by 12 April and had completely melted by 16 April, a week earlier than W-9.

Isotopic and chemical evolution of the snowpack

The heterogeneity of $\delta^{18}\text{O}$ in the layered snowpack was initially determined by the variability of precipitation $\delta^{18}\text{O}$, which ranged from -24 to -6‰ during the study period (Fig. 2). From December through mid-February, precipitation $\delta^{18}\text{O}$ was generally -20‰ or less, with seasonal minimum values in January. After mid-February, $\delta^{18}\text{O}$ of precipitation was more variable, but generally increased from -15 to about -10‰ . The $\delta^{18}\text{O}$ of rain events was nearly always several per mill heavier than snow events in the same time period. The effect of the late January rain-on-snow

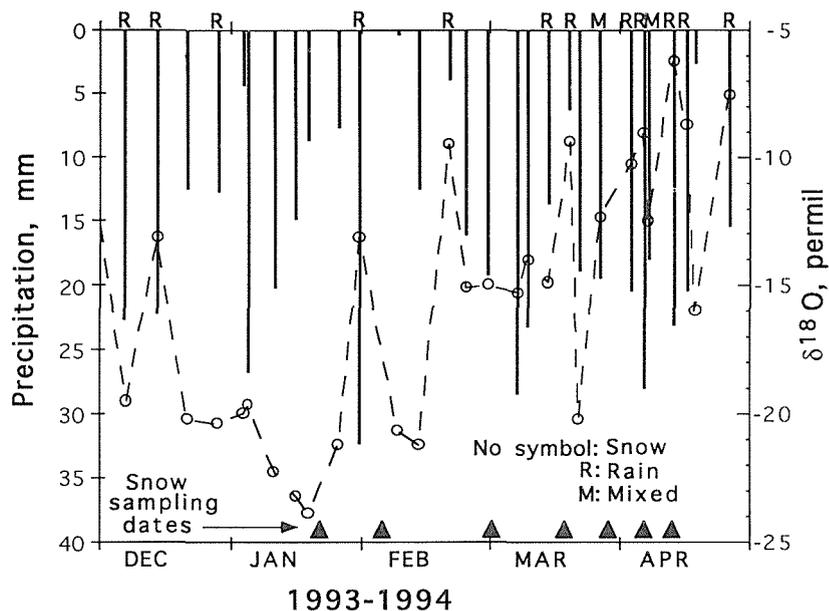


Fig. 2 Amounts and $\delta^{18}\text{O}$ of precipitation events. Open circles are $\delta^{18}\text{O}$ values; vertical bars are precipitation amount.

event, which had a relatively heavy $\delta^{18}\text{O}$ of -13‰ , is reflected in the isotopic depletion of the upper part of the 4 February snow profiles (Fig. 1(a)).

The winter was divided into an accumulation period and an ablation period (Fig. 1); peak accumulation on 18 March was used to divide the two periods. Snow accumulated consistently throughout January and February, which were relatively free of rain and warm weather with the notable exception of a major rain storm on 28 January. The rain appeared to infiltrate less than 20 cm below the snow surface; below that depth, the isotopic profile maintained a pattern that was nearly identical to the pre-rain condition. Through peak accumulation on 18 March, the two profiles changed little in the lower part of the snowpack, but the isotopic pattern in the upper part of the snowpack reflected the accumulation of increasingly heavy snows and small rain events. The 1 March snow pit at W-9 had an anomalously low depth because the snow pit for that date was on a hummock; despite this "shortening" of the profile, the $\delta^{18}\text{O}$ pattern with depth was similar to that on 4 February.

After 18 March, a series of isotopically enriched rain storms and warmer temperatures caused water to flow through the pack, changing its isotopic profile. The infiltrating rainfall, as well as fractionation during the melting process, resulted in an increasingly heavy pack. Although the bottom 20 cm of each pack changed by only 2‰ , mid-pack positions changed by as much as 6‰ . By the last sampling date, the 11 cm of snow remaining at W-2 had a relatively enriched $\delta^{18}\text{O}$ of -18.5‰ .

The chemical evolution of the snowpack was affected by highly variable inputs in precipitation; nitrate concentrations in rain and snow ranged from 5 to $95\ \mu\text{eq l}^{-1}$. Relative to the pattern of $\delta^{18}\text{O}$, the pattern of nitrate concentrations in the snowpack during the accumulation period changed more rapidly (Fig. 1(a)). This was not an unexpected result, given that a large fraction of the solutes move with a small fraction

of the initial meltwater. Prior to the 28 January rain-on-snow event, the snowpack surface was high in nitrate. On 25 January, a snowstorm of 0.75 cm water content and $85 \mu\text{eq l}^{-1}$ nitrate accumulated on a surface layer that already had $96 \mu\text{eq l}^{-1}$ nitrate on 21 January (Fig. 1(a)). As seen in the 4 February profile (Fig. 1(a)), the downward movement of this nitrate pulse was impeded by the ice layer just below the snow surface. At W-2, this ice layer was less well-developed, and elevated nitrate was observed to a depth of 15 cm below the snowpack surface. By peak accumulation, the nitrate pulse had moved downward in both profiles, despite little melting and only two small rain events. Although some of the downward movement may have resulted from settling, the progressive damping of the nitrate peak associated with this pulse suggests that nitrate was redistributed.

As flow through the snowpack increased during the ablation period, nitrate appeared to move more freely through the pack, probably as a result of weakening ice layers. High-nitrate rains on 3 April ($44 \mu\text{eq l}^{-1}$), 6 April ($34 \mu\text{eq l}^{-1}$), and 7 April ($28 \mu\text{eq l}^{-1}$), each with greater than 1.5 cm water content, actually lowered nitrate concentrations in the snow profiles (Fig. 1(b)). The flushing of nitrate was more complete at W-2, possibly because the snowpack was thinner and riper, and because there was less leaching of nitrate from organic debris at this open site. The general progressive decrease of nitrate concentrations in the snow profiles during the ablation period is field evidence of preferential elution of nitrate with meltwater discharge from the snowpack (e.g. Johannessen & Henriksen, 1978).

Comparison of $\delta^{18}\text{O}$ and chemistry of snow cores and snow layers

Addition of plant litter to the snowpack is a natural part of the snow accumulation process. This material influences microbial activity within the snowpack and affects the chemical composition of the meltwater. However, the vegetative material has a greater impact on relatively dilute water chemistry of snow samples collected and melted for chemical analysis than it does on snowpack meltwater because of the greater contact time in the melted snow prior to filtration in the laboratory. If solute leaching from organic material occurs, spatial variations in the distribution of organic material may give rise to spatial variations in snowpack chemistry.

To evaluate spatial variations in snowpack chemistry, we compared solute loadings in the snowpack, as determined by compositing the loads in individual layers, to that determined directly on the snow core taken nearby. This comparison was repeated for

Table 1 Maximum and median percent differences, on 6 sampling dates at each site, between snowpack solute loads determined from (a) single snow cores and (b) sum of loads in individual snow layers, expressed as percentage of lesser number; $\delta^{18}\text{O}$ column gives absolute difference (in ‰).

	Ca	Mg	Na	K	Cl	NO ₃	SO ₄	$\delta^{18}\text{O}$
W-2								
Maximum	238.7	287.1	124.7	418.0	25.3	127.0	106.9	2.6
Median	20.1	57.4	36.5	141.5	7.4	5.5	16.4	0.9
W-9								
Maximum	53.3	257.5	60.5	536.4	65.3	43.5	80.9	0.5
Median	27.5	55.3	33.7	64.9	10.9	7.1	3.7	0.2

each sampling date at each site, Similar comparisons were made for $\delta^{18}\text{O}$; because $\delta^{18}\text{O}$ is unaffected by vegetative matter, agreement of $\delta^{18}\text{O}$ cores and layer composites can be used as an indicator of representative sampling. Maximum and median differences between the two determinations for each solute and site suggest that chemical alteration by organic matter is a pervasive problem at both sites (Table 1). The relatively close agreement of the $\delta^{18}\text{O}$ determinations, particularly at the forested site, tends to dismiss sampling nonrepresentativeness as the cause of the discrepancies. Indications of an organic matter effect include the large magnitude of the maximum discrepancy for most solutes, the large difference between maximum and median discrepancies, and the generally high discrepancies for K, which is readily leached from organic material.

Relation of snowpack evolution to snowmelt and stream chemistry

Isotopic, chemical, and hydrometric evidence from the 1993 snowmelt at Sleepers River suggest that stream water during snowmelt is dominated by groundwater discharge at W-9, and by snowmelt water at W-2 (Shanley *et al.*, in review). The present study was based on the need for a better understanding of the mechanisms by which snowmelt water is delivered to the stream. At W-9, initial meltwater was isotopically depleted relative to the snow core (Fig. 3(a)), because of fractionation during the melt process. The apparent isotopic enrichment of the later meltwater was probably caused in part by rain enriched in ^{18}O (Fig. 2) because the meltwater lysimeters also collect drainage from rain-on-snow events. The meltwater was at all times isotopically lighter than stream water, which had little isotopic response to the strong temporal $\delta^{18}\text{O}$ pattern in meltwater. As in the 1993 melt, it appears that the stream was dominated by groundwater discharge, and most of the snowmelt water infiltrated and recharged the shallow aquifer. A possible exception was the period in mid-April when the snowmelt isotopic signal approached that of the stream, making an isotopic separation more difficult; this was also the time of the seasonal maximum flow.

At W-2, the snowpack was thinner and it ripened earlier than the pack at W-9. Thus, late-season rainfall flowed more readily through the pack. The isotopic enrichment of the rainfall is reflected in $\delta^{18}\text{O}$ values of the lysimeter outflow that were consistently enriched relative to the bulk snowpack. Meltwater $\delta^{18}\text{O}$ values fluctuated as outflow was

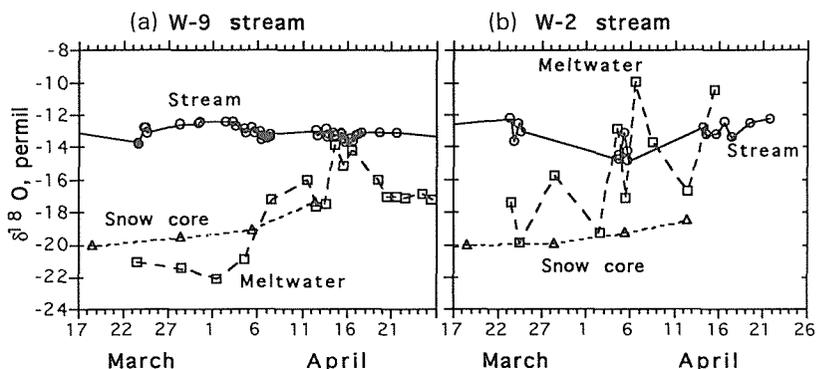


Fig. 3 $\delta^{18}\text{O}$ of snow core, meltwater, and stream water at Sleepers River during the 1994 snowmelt. (a) W-9; (b) W-2.

driven alternately by rain-on-snow and thermal snowmelt. For much of this period, meltwater $\delta^{18}\text{O}$ overlapped groundwater $\delta^{18}\text{O}$, which was assumed to have a $\delta^{18}\text{O}$ near pre-melt streamflow, making hydrograph separations impossible. However, stream-water $\delta^{18}\text{O}$ at W-2 responded discernably toward the isotopically lighter (in general) $\delta^{18}\text{O}$ in snowmelt. The stream at W-2 thus appeared to contain a much higher proportion of snowmelt than the stream at W-9, which had little isotopic response. Unnikrishna *et al.* (1993) found that the $\delta^{18}\text{O}$ of meltwater discharge corresponded to the $\delta^{18}\text{O}$ of the bottom 10 cm of a relatively homogenous California snowpack. At both sites in the present study, the bottom of the pack maintained a fairly constant $\delta^{18}\text{O}$ during the ablation phase (-21‰ at W-9 and -18‰ at W-2) (Fig. 1(b)). These values were at the lower limit of meltwater $\delta^{18}\text{O}$ (Fig. 3), which was commonly enriched, probably from isotopically heavy rainfall.

The contrasting $\delta^{18}\text{O}$ behaviors of W-2 and W-9 provide a good example of the need for sampling temporal changes in the $\delta^{18}\text{O}$ values of the new water end-member for hydrograph separations. At W-9, the interstorm and intrastorm variability in rain $\delta^{18}\text{O}$ is damped by snowmelt throughout the melt. Because the temporal changes in the $\delta^{18}\text{O}$ of meltwater and meltwater-plus-rain are gradual, one can safely interpolate between known values and model the response with two components. Late in the melt period at W-2, in contrast, the isotopic variability of rain is no longer damped by the composition of melting snow. Thus, the new water end-member can no longer be modeled as a gradual change in the compositions of lysimeter outflow samples collected several days apart, and simple two-component hydrograph separations become unrealistic.

Considerable controversy persists over the relative contributions of snow nitrate and soil nitrate to surface water nitrate increases during spring runoff (Rascher *et al.*, 1987). Preliminary results from a pilot study at Sleepers River in 1994 suggests that the dominant source of nitrate in stream water is from pre-event soil water, not nitrate from the snowpack (Kendall *et al.*, 1995, this volume). Because nitrate concentrations in stream water vary widely during snowmelt, it may be possible to relate nitrate behavior in stream water to nitrate release from the snowpack. Evaluation of nitrate contributions from the snowpack to stream water are not straightforward because meltwater may acquire soil nitrate as it flows through the shallow soil zone. Meltwater and soil nitrate contributions would appear in stream water at about the same time.

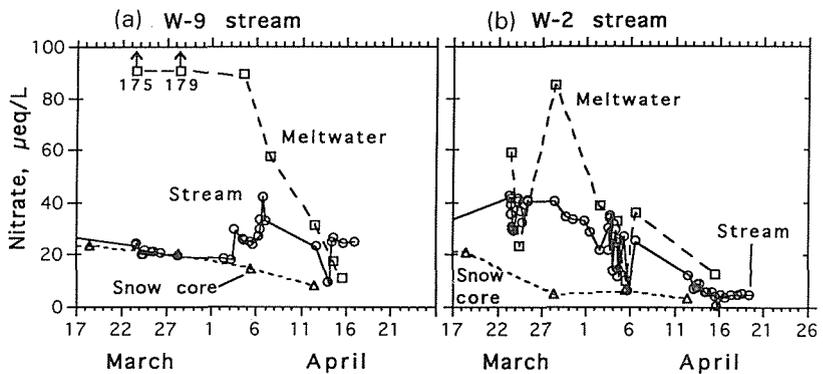


Fig. 4 Nitrate concentrations of snow core, meltwater, and stream water at Sleepers River during the 1994 snowmelt. (a) W-9; (b) W-2.

Stream-water nitrate at W-9 increased sharply in association with discharge increases on three occasions during the runoff period (Fig. 4(a)). It was not possible in this study to determine whether the source of nitrate in stream water was soil nitrate or snowpack nitrate. There appeared to be ample nitrate in meltwater to account for the high nitrate concentrations in stream water. Meltwater nitrate concentrations were several times higher than snowpack nitrate concentrations. High nitrate concentrations in the initial meltwater were consistent with concentration factors of near 10 observed by Marsh & Pomeroy (1994). Meltwater also reflects late-season rain concentrations of nitrate and ammonium, which were higher than those in the bulk snowpack. However, total loading of nitrate in meltwater could not be determined because meltwater volume data were insufficient. Thus, soil nitrate contributions cannot be ruled out.

Meltwater nitrate concentrations at W-2 were likewise higher than snowpack nitrate concentrations (Fig. 4(b)). However, variations in nitrate concentrations in the W-2 stream were well-correlated to those in meltwater, suggesting that the snowpack may be the dominant source of nitrate to the stream. This conclusion is consistent with the $\delta^{18}\text{O}$ data which suggest that stream water at W-2 is dominated by new water inputs during snowmelt.

CONCLUSIONS

Field evidence supports previous observations that preferential ion elution and preferential release of isotopically depleted meltwater due to isotopic fractionation are important to the evolution of a layered snowpack. However, the effect of rain-on-snow events with significantly enriched $\delta^{18}\text{O}$ composition, and contamination from organic debris may also have strong influences on the isotopic and chemical evolution of the snowpack. The timing of water and solute release is further affected by the complex morphology of the layered pack. Water and nitrate moved through the snowpack more readily as the pack ripened.

The results of this study underscore the importance of using the $\delta^{18}\text{O}$ values of snowmelt, not snow cores, for the new water end-member for hydrograph separations. The results from these two sites demonstrate that land cover and elevation have a significant effect on snowpack accumulation and ablation, chemical composition, morphologic evolution, and meltwater $\delta^{18}\text{O}$ values. Because of the rapid changes in the $\delta^{18}\text{O}$ of snowmelt late in the melt period, differences in the timing of snowmelt at different elevations can critically affect the accuracy of hydrograph separations. Hence, several melt lysimeters are necessary in catchments with highly variable topography and/or land cover.

Acknowledgments The authors are grateful for laboratory assistance from Mark Olson, Deb Horan-Ross, and Trish Lincoln of the U.S. Geological Survey Water Quality Laboratory in Albany, New York, and Alan Hewitt and Pat Schumacher of the Cold Regions Research and Engineering Laboratory in Hanover, New Hampshire.

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