163: Hydrochemical Processes in Snow-covered Basins

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This article reviews several aspects of snow hydrochemistry: the chemistry of snowfall including chemical incorporation in snowfall and snowfall chemistry variability, the chemistry of cold, dry snowcovers including snow redistribution, snow–atmosphere chemical exchange and in-pack chemical transformations, the chemistry of wet and melting snowcovers including solute leaching, particulate interactions and microbial activity, and snow-covered basin hydrochemistry with an emphasis on nutrient chemistry. The emphasis is on the processes of chemical transformation in seasonal snowpacks and meltwaters with strong attention to the broad ecosystem view of snow chemistry rather than solely focusing on acidification effects from snowmelt. The seasonal snowcover is shown to be a dynamic hydrochemical system with strong ecological interactions. Besides wet deposition by snowfall and rain, the processes of wind redistribution, dry deposition, volatilization, crystal metamorphism, photolysis, microbial uptake and release, solute elution, and meltwater movement strongly affect the chemistry of both the snowpack and meltwaters. Snowmelt chemistry alone is rarely directly responsible for major chemical fluctuations in water bodies, but meltwater has an important role in transporting ions from soils and organic material to water bodies.

INTRODUCTION

The seasonal snowcover plays a unique role in basin hydrochemistry by collecting and transforming chemicals over the snow accumulation season and then releasing them suddenly during melt. Chemicals in the snowcover are derived from dry and wet deposition and are preferentially flushed out, early in the melt season. When industrial pollutants are scavenged and deposited to snowcover, the resulting meltwaters provide a low pH contribution to aquatic and soil chemistry (Hendershot *et al.*, 1992; Galloway *et al.*, 1987). Tranter and Jones (2001) organized the processes, which influence the chemical composition of snow and meltwaters, into those involving

1. heat and mass fluxes that occur during sublimation and melting; and
2. chemical transformations.

During sublimation and melting, chemical species are considered conservative (excluding migration) in that they are not transformed. The physical properties of chemical species (e.g. solubility, vapor pressure) are therefore important in how these processes alter the chemical load of, and release from, the snow cover (Brimblecombe and Shooter, 1991). Chemical transformations occur by chemical reactions such as oxidation (Bales *et al.*, 1987) and photolysis (Beine *et al.*, 2002), or from microbiological activity (Hoham *et al.*, 1989). This review will consider hydrochemical processes in snow from snowfall formation to infiltration and runoff.

The Chemistry of Snowfall

Wet deposition to snowcovers primarily occurs through snowfall. The reader is referred to sections on rainfall chemistry for a discussion of the chemical inputs that can derive from rain-on-snow events (Stumm and Morgan, 1996).

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Chemical Incorporation in Solid Precipitation

Solid precipitation particle formation and fall through the atmosphere incorporate atmospheric chemicals via three main processes:

1. imprisonment during the initial formation of ice crystals;
2. capture of gases, aerosol, and larger particulates within clouds; and
3. scavenging of these materials below the cloud layers during snowfall (Barrie, 1991).

Cloud water droplets contain solute as a result of aerosol scavenging, and the diffusion of atmospheric gases into solution. The soluble species are mainly NH_4, SO_4, NO_3, Ca, K, and Mg, derived from natural and anthropogenic emissions, in addition to Na and Cl from sea-salt aerosol. In addition to solute, droplets contain sea-salt aerosols, particulate organic debris, and/or fine particulate clays (Kamai, 1976) that can form ice nuclei for freezing. The chemical content of the center of falling snow grains is often characterized by these compounds. During the droplet freezing process, however, most solutes are rejected to the outer edges of the crystal as they do not fit well into the ice crystal lattice (Colbeck, 1987).

Ice crystals then grow by vapor transfer from supercooled water droplets because of small differences in saturation vapor pressure over ice and water. Very little solute is transported by this process to the growing ice crystals (Hewitt and Cragin, 1994), however, small amounts of HNO_3 and HCl are absorbed. Direct collision between growing crystals and supercooled droplets results in rimed crystals and graupel, which contain relatively high solute concentrations compared to crystals formed mainly by vapor transfer (Cerling and Alexander, 1987).

Further scavenging of aerosols and other particulates occurs below clouds by adsorption and impact; snowfall more efficiently scavenges particulates than does rain because of its higher surface area to mass ratio, and lower terminal fall velocity (Raynor and Haynes, 1983; Nicholson *et al.*, 1991).

Relationships between crystal form and chemistry have been found because of the association between genesis and form, and the strong differences in chemical incorporation during various genetic processes. Lamb *et al.* (1986) found that the highest concentrations of solute were associated with the smallest unrimed crystals. Borys *et al.* (1983) showed the solute content of snow crystals to increase with the degree of riming. Hewitt and Cragin (1994) found that dendritic and stellar plates had similar chemical content for most species, except for Cl, which showed much higher concentrations in stellar plates than dendritic crystals.

Variability of Snowfall Composition

Snowfall chemical composition depends on factors such as the air mass origin, altitude, and meteorological conditions (Colin *et al.*, 1989; Davies *et al.*, 1992). Maritime air masses will give rise to snow containing mostly Na and Cl (Tranter *et al.*, 1986), while polluted air masses from industrial areas will deposit snow that is highly acidic because of the presence of strong acid anions (NO_3, SO_4) from fossil fuel combustion (Davies *et al.*, 1984; Landsberger *et al.*, 1989).

The spatial variability of snowfall chemistry shows length scales from meters to hundreds of kilometers (Tranter *et al.*, 1987; Pomeroy *et al.*, 1995; Turk *et al.*, 2001; de Caritat *et al.*, 2005), reflecting factors such as the proximity to pollution or ocean sources, the back trajectory of the air masses associated with the snowfall, and wind mixing of different snowfalls on the ground. Generally, snowfall at high altitudes contains lower chemical concentrations than at lower altitudes because of the shorter air column for the falling snow crystals to scavenge from. In addition, chemical concentrations during a snowfall event often decrease exponentially with time, as the store of species available for scavenging in the atmosphere depletes with cumulative scavenging. As a result, persistent storm tracks (e.g. lake effect, coastal) have characteristic spatial trends in snowfall chemistry. For instance, as air masses rise up the windward side of mountains, they generate heavy orographic snowfall. This snowfall progressively depletes aerosols over the increasing distance, resulting in more dilute snowfall chemistry with increasing altitude (Lyons *et al.*, 1991). Postdepositional changes and changes due to redistribution of snow are dealt with in the next section.

Chemistry of Cold, Dry Snowcover

Snowcovers at temperatures below 0 °C are defined as "cold", and because of low liquid water contents are termed "dry". The temporal and spatial variation in the chemical composition of snowfall and subsequent redistribution processes usually produce a snowcover that is chemically heterogeneous. The main processes of transformation (see Figure 1) are

1. redistribution by wind and vegetation;
2. surface-exchange at the snow-atmosphere interface (dry deposition and volatilization);
3. surface and subsurface chemical reactions;
4. snow-grain metamorphism within the pack; and
5. basal-exchange processes at the snow-soil interface (gaseous emissions from soil).

Snow Redistribution Processes

Snow is redistributed by wind via blowing snow transport (Pomeroy *et al.*, 1991), and by vegetation via snow

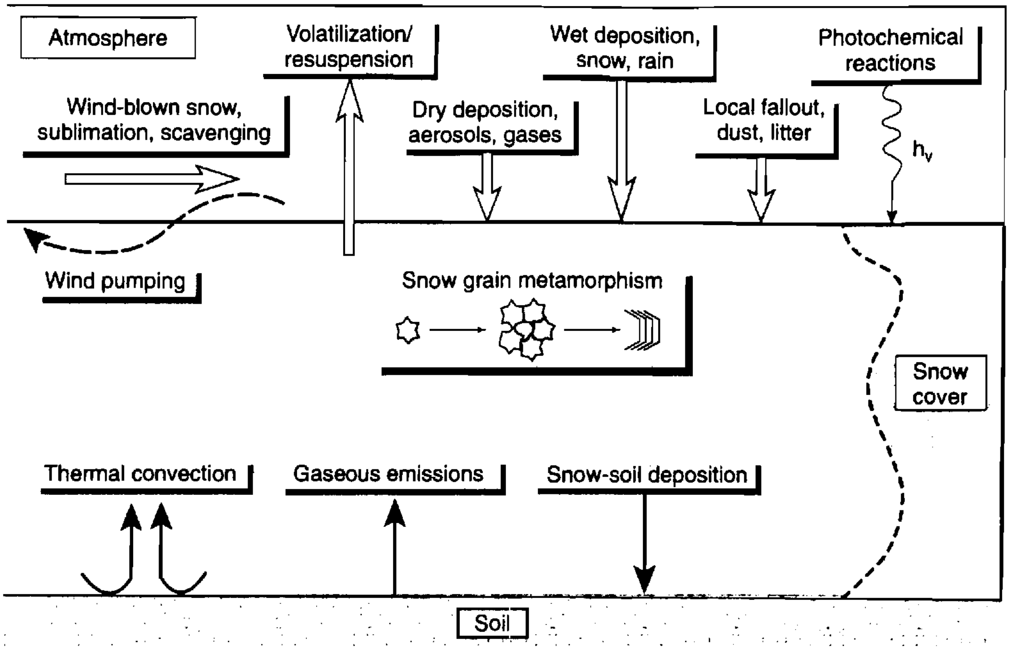

Figure 1 The main physical and chemical processes that influence the chemical composition of cold, dry snow cover during the accumulation season (After Tranter and Jones, 2001)

interception (Pomeroy *et al.*, 1999). Wind transport has the potential to change the chemical composition of snow due to three main physical processes, namely, sublimation of water vapor, scavenging of aerosols and gases from the atmosphere, and volatilization (Pomeroy and Jones, 1996). Direct wind redistribution of snow chemicals can move chemical species between basins, and both transport and sublimation increase the spatial variability of snow chemistry. In the Cairngorm Mountains of Scotland, Tranter *et al.* (1987) found that snowfall chemical concentrations had coefficients of variation (CV) from 0.03 to 0.04. However, CVs for ion concentration in wind redistributed snowcovers at this site varied from 0.1 to 0.74 (Pomeroy *et al.*, 2000). At the arctic treeline in NW Canada, Pomeroy *et al.* (1995) found that blowing snow redistribution was associated with a fivefold difference in chemical loading in snow within a 70 km^2 basin; this greatly exceeded the differences due to ion concentration for which the CVs ranged from only 0.06 to 0.12.

Interception by evergreen canopies can store over half the cumulative snowfall in midwinter (Pomeroy and Gray, 1995). Where chemical species are conserved, redistribution from trees occurs at scales of only a few meters and is generally unimportant. Dry deposition and volatilization are also affected by snow interception, and are discussed in the following section. Sublimation of intercepted snow increases the concentration of conserved ionic species up to sixfold, according to the loss of ice from intercepted snow clumps (Pomeroy *et al.*, 1999).

Processes at the Atmosphere-snow Surface Interface

Dry Deposition

Dry deposition is the direct deposition of chemical species from the atmosphere to the snow surface (Cadle, 1991). Aerosols and particulates may be directly deposited, while gaseous species may be adsorbed (Conklin, 1991). Because the aerodynamic surface roughness of snow is low (Cadle *et al.*, 1985), and liquid water layers are small to nonexistent in cold snow (Choi *et al.*, 2000), dry deposition to snow cover is much lower than to surfaces without snow or to forest canopies, and is small in relation to wet deposition from snowfall.

Bales *et al.* (1987) found that dry deposition rates to new snow were higher than those to old snow as a result of a reduction in the area of crystal surfaces during the metamorphism of snow. The importance of crystal form in dry deposition has also been reported by Ibrahim *et al.* (1983), who suggested that the interception of aerosols by ice needles in relatively fresh snow contributed significantly to the measured rates of dry deposition. Albert and Shultz (2002) show the effect of within-snowpack ventilation via diffusion and advection in enhancing dry deposition to snow.

Cadle *et al.* (1985) estimated deposition rates for HNO_3 to be one order of magnitude larger than those for SO_2. The difference was attributed to the relative solubility of the two gases in the liquid layers around the crystals, and also to other factors such as relative diffusion rates into the ice lattice, and the rate of oxidation of SO_2 to SO_4 at the air–crystal interface (Bales *et al.*, 1987; Bales, 1991). The rate of deposition of HNO_3 is much higher to wet snow than to cold, dry snow (Cadle, 1991). Field studies in North America suggest that dry deposition contributes approximately 20–25% of the chemical composition of SO_4 and NO_3 in snowcovers (Cadle and Dasch, 1987; Barrie and Vet, 1984; Cadle, 1991).

Snow covers under forest canopies may receive greater contributions of dry deposition. Pomeroy *et al.* (1999) showed greater dry deposition of aerosols to intercepted snow, and subsequent unloaded snow in forest canopies. Dry deposition of gases and aerosols to forests is higher than to open snowfields because of enhanced aerodynamic roughness and absorptive needle/bark surfaces (Höfken *et al.*, 1983; Dasch, 1987). Species deposited to the canopy are subsequently redistributed to the snowcover via intercepted snow unloading, or rainfall interception and drip.

Aeolian dust and other particulate matter are continuously being deposited on snow covers. The result of dust deposition is usually a reduction in the acidity of the snow (Sequeira, 1991), particularly during melt periods (Clow and Ingersoll, 1994; Delmas *et al.*, 1996). Pomeroy *et al.* (1991) found enhancements of aerosol concentration in postdepositional and in-transit wind-blown snow during periods in which blowing snow particles developed strong electrical charges, and suggested that electrophoresis could attract small aerosols to blowing snow particles before deposition. The net effect of changes in aerodynamic roughness on dry deposition of gaseous species and distance from source of aerosols can be difficult to distinguish. An example of loadings of ions to basins with and without trees is shown in Figure 2.

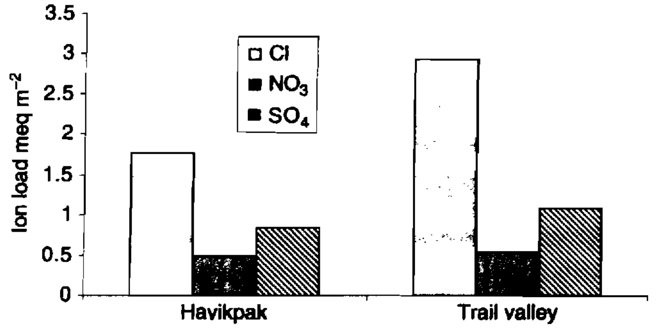

Figure 2 Ion loadings in snow to two basins in NW Canada; Havikpak Creek is sparsely forested and further away from the Arctic Ocean, Trail Valley Creek is dominated by tundra and 50 km closer to the sea. Differences in dry deposition of SO_4 and Cl are evident and because of the increased roughness of the Havikpak forests, and to the different availability of aerosol from the ocean (After Pomeroy *et al.*, 1995)

Snow will also accumulate nutrients by the deposition of biological debris, either as fallout from above the snow surface, or by direct incorporation into the snowpack. In forests, much of the deposition arises as litterfall from the canopy (Jones and Debois, 1987) and mammalian excrement (Jones, 1991). Invertebrate fallout from wind-borne arthropods and winged invertebrates may also contribute nutrients to snow, while vertebrate and invertebrate activity under and within the snow cover transforms and redistributes nutrients from the soil, upwards into the snow matrix. Jones (1991) attempted to calculate nutrient inputs to snow from different vertebrates by considering the spatial distribution of population densities, and the amount and chemical composition of animal excrement. Persistent spatial distributions of excrement were important to basin-scale estimates of N deposition. In the case of animals that herd (e.g. deer), deposition rates may be up to 200 g N ha^{-1} day^{-1} in areas of congregation. Solitary mammals, such as moose and hares, deposit only up to 2.5 g N ha^{-1} day^{-1}.

Volatilization

The dry deposition of aerosols to snow is often considered to be irreversible. However, some species may volatilize and be lost back into the atmosphere. Postdepositional loss of NO_3 has been observed in surface snow (Neubauer and Heumann, 1988; Beine *et al.*, 2002), and sublimating intercepted and blowing snow (Pomeroy *et al.*, 1991; Pomeroy and Jones, 1996; Pomeroy *et al.*, 1999). Volatilization of NO_3 in redistributed snow was proportional to ice sublimation losses over the winter, which is on the order of 35% in the boreal forest, 20% in arctic tundra, and 20–40% in the northern steppes. Figure 3 shows ion concentration enrichment as a function of sublimation loss of snow mass for NO_3, SO_4, and Cl in a boreal forest. SO_4 and Cl are conserved, as snow mass is decreased by sublimation; the loss of NO_3 is roughly proportional to the loss of snow mass, suggesting an association between volatilization and sublimation. These results are consistent with the observations of Stottlemyer and Troendle (1999) who observed increases in NO_3 in snowpacks and meltwater runoff, when evergreen forest basins were clear-cut in the Rocky Mountains, Colorado.

Oxidation and Photochemical Reactions in Surface Snowcover

Oxidation of certain species by atmospheric oxidants may take place on cold snow-grain surfaces, if a liquid film is present (Conklin and Bales, 1993). Bales (1991) has modeled the chemical oxidation of S(IV), SO_2, to S(VI), SO_3/SO_4, on the basis of the known oxidation rates by H_2O_2, O_3, and O_2. Whilst of great interest to ice core interpretations, this also has relevance to understanding S uptake in snow-covered catchments.

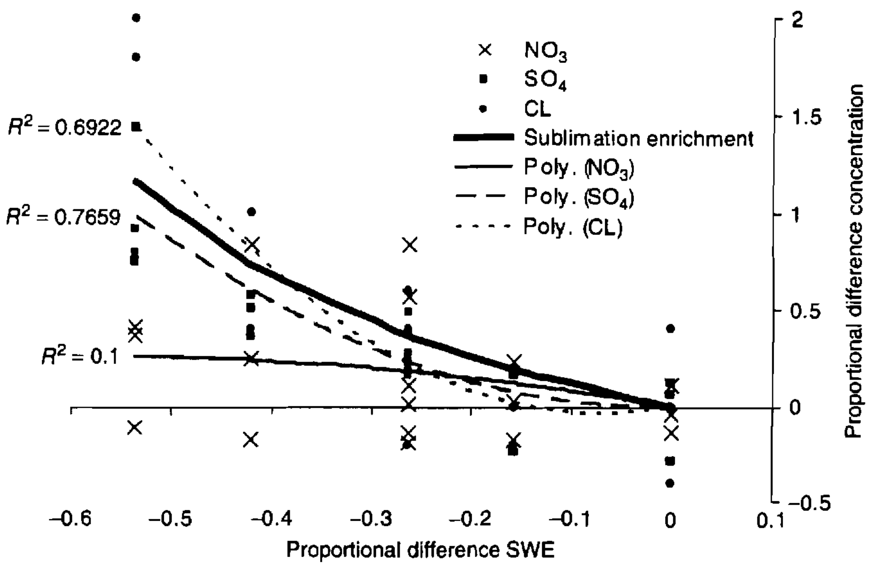

Figure 3 Enrichment of ion concentration because of sublimation of intercepted snow in the boreal forest. SO_4 and Cl follow the expected enrichment assuming conservation of ion, whilst NO_3 is depleted during sublimation. Snow mass is shown as snow water equivalent (SWE), and both SWE and ion concentration are referenced to levels that in a clearing experienced neither melt nor snow redistribution (After Pomeroy *et al.*, 1999)

Sigg *et al.* (1987) measured the decrease in concentrations of H_2O_2 in surface alpine snows, and proposed that photolysis was the primary mechanism. Neubauer and Heumann (1988) suggested that the apparent loss of NO_3 from Antarctic snow was due either to the photodegradation of HNO_3 to NO_2 by solar radiation, and/or to the volatilization of HNO_3 from snow during metamorphism, but were unable to distinguish between the two mechanisms. Beine *et al.* (2002) found that photolysis on a high Arctic snowpack caused HNO_3 transport to the snow surface.

In-pack Processes

Metamorphism

The processes of snow metamorphism are described in **Chapter 160, Energy Balance and Thermophysical Processes in Snowpacks, Volume 4**. It is currently accepted that the solute becomes redistributed to, and concentrated on, the snow-grain surfaces or in snow particle bonds during dry snow metamorphism, although direct observation of this process has not yet been made. The net effect of weak temperature gradient metamorphism is to concentrate the solute onto or near the surfaces of ice crystals (Bales, 1991; Colbeck, 1987; Davis, 1991). The solute may be located in a "quasi-liquid" surface layer, as discrete aerosol or as concentrated, "doped ice" pockets (Davis, 1991). Strong temperature or kinetic metamorphism is also accompanied by loss of ions. Ion losses are likely due to some form of transport, either to the base of the pack or to adjacent snow strata and the atmosphere. Laberge and Jones (1991) found that SO_4 was lost during depth hoar formation. In contrast, Pomeroy *et al.* (1993) found that SO_4 and Cl concentrations in depth hoar increased in proportion to the overall loss of water vapor from the depth hoar to adjacent snow layers. However, NO_3 concentrations remained approximately constant, indicating a concomitant loss of the species.

Chemistry of Wet Snow and Snow-meltwater Systems

The percolation of meltwaters through the snowcover (*see* **Chapter 161, Water Flow Through Snow and Firn, Volume 4**) causes the chemical composition of both the snow matrix and the meltwaters to change. The concentration and distribution of solutes in the snow-meltwater system is controlled by a wide variety of physical and biological processes (see Figure 4). These processes include

1. solute leaching from snow grains;
2. meltwater-particulate interactions; and
3. microbiological activity.

In addition, snow-atmosphere exchange is another factor as dry deposition rates of certain species (e.g. SO_2, HNO_3, HCl) to wet snow crystals increase significantly because of their solubility in water (Cadle, 1991). Rain on snow will also influence the chemistry of meltwaters due to its own chemical composition (Tranter *et al.*, 1992).

Solute Leaching

Fractionation of solute species between snow grains and meltwater occurs because of leaching of the melting snow

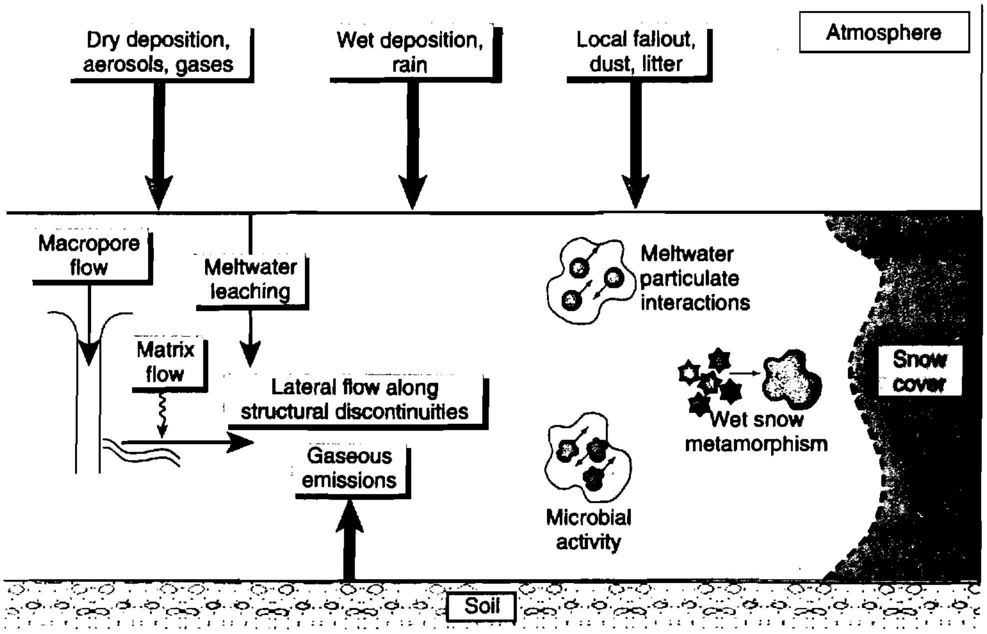

Figure 4 The main physical, chemical, and biological processes that influence the chemical composition of snow cover during melt (After Tranter and Jones, 2001)

grains. The result is that the meltwater front becomes progressively more concentrated as it moves through the pack (Johannessen and Henriksen, 1978; Colbeck, 1981). The degree of fractionation of any solute species, x, between snow and meltwater can be described by a nondimensional "concentration" factor, CF,

$$\mathrm{CF} = \frac{C_m}{C_p} \qquad (1)$$

where C_m is the ion concentration in any meltwater fraction, and C_p is the concentration in the parent snow prior to melt. Values of CF during the initial stages of meltwater discharge may range from 1 to 50, but a more typical range is 2 to 7 (Tranter, 1991). CF decreases with increasing melt and cumulative leaching to values of <0.1 in the final meltwaters. The efficiency of meltwater leaching (i.e. higher values of CF per volume of initial meltwater discharge) depends on the distribution of solute in snow grains, snowpack, and meltwater flow paths.

Wet snow metamorphism in the melt period is more rapid than dry snow metamorphism. It causes large grains to grow at the expense of small grains and solute to diffuse from grain boundaries into meltwater (Davis, 1991). The solute scavenging that results in fractionation is related to the rate of melt (Tsiouris *et al.*, 1985; Tranter *et al.*, 1988b) because it is affected by diffusion rates and the duration of snow-meltwater interaction. For example, Marsh and Webb (1979) reported the approximate doubling of initial snowmelt concentrations with the doubling of snow depth. Conversely, at high melt rates, solute scavenging is minimized. Diurnal changes in melt rate affect the CF of meltwaters, with the highest concentrations being found in the morning and evening, or during periods of shading, when melt rates are lowest.

Because of solute scavenging, meltwater flowing rapidly through snow macropores or "flow fingers" is more dilute than melt flowing through the snow matrix. The effects of heterogeneous flow on the spatial variability of fractionation are illustrated in Figure 5, where the CF is shown for two flow paths, one with the lowest (matrix) and one with the highest (macropore) measured flow rate (Marsh and Pomeroy, 1999). The CFs of both flow paths gradually converged over time, until all flow paths had similar values.

The mesoscale distribution of solute in snow cover will also affect the concentration of meltwaters. Discrete snowfalls or redistribution events cause snow strata to have differing composition. Solute-rich bands can arise from the exclusion of solute from ice lenses formed by the refreezing of meltwater, or rainwater in cold snow. The result of several diurnal melt-freeze cycles is often to increase the ionic concentrations in the first meltwaters issuing from the snowpack (Bales *et al.*, 1989; Williams and Melack, 1993). Both laboratory and field experiments have shown that solute-rich layers give rise to more concentrated meltwaters (Colbeck, 1981; Tranter *et al.*, 1986; Marsh and Pomeroy, 1999).

Modeling solute leaching and meltwater composition is extremely difficult because of uncertain processes, variable location of solutes in snow, and complex meltwater

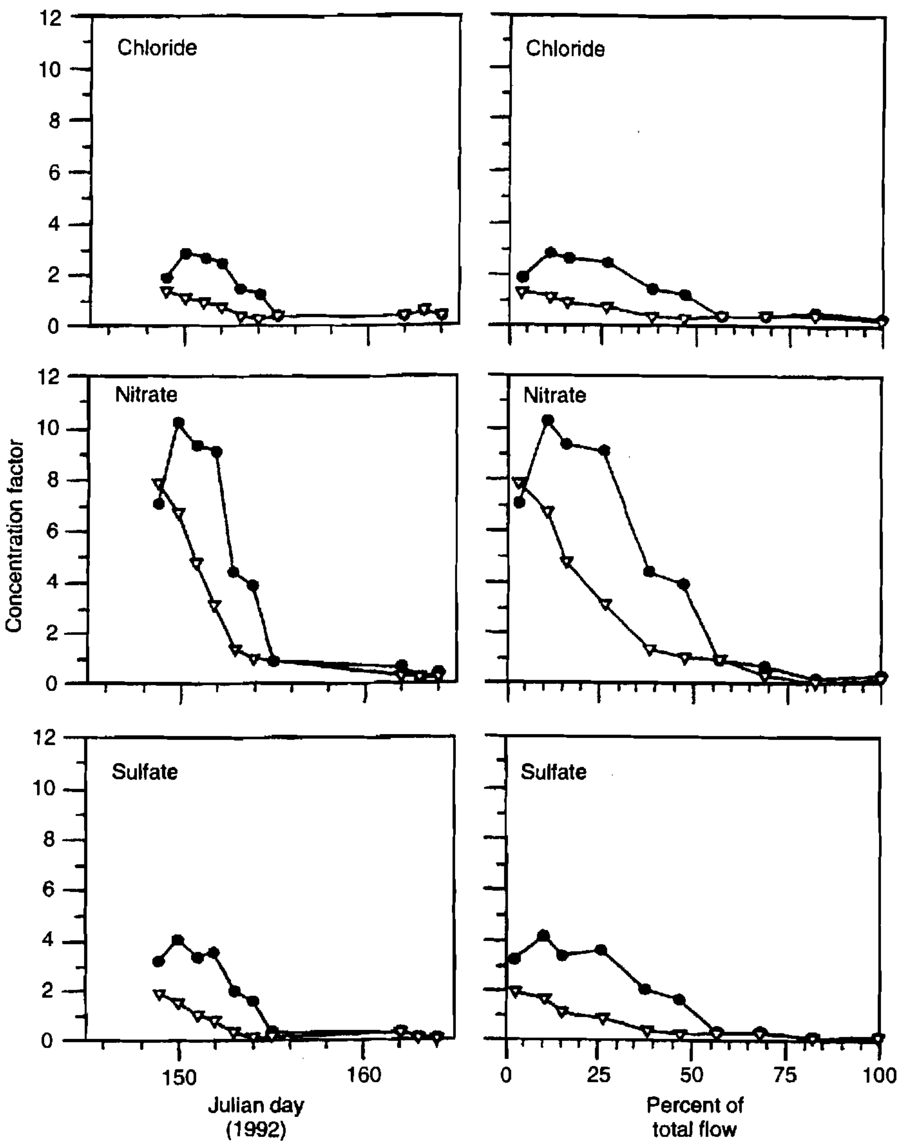

Figure 5 The impact of flow rate on the concentration factor of S042-, S03- and Cl- in snowmelt. The data is collected from a multiple compartment lysimeter under a tundra snowpack that samples both high and low meltwater flow zones of the melting snowpack. Open symbols and closed symbols denote the concentration factors from high and low flow chambers respectively

dynamics. Many early elution models were based on bulk transport, which in turn was based on snow depth and a bulk leaching coefficient (Stein *et al.*, 1986). The leaching coefficients were based on first-order removal of the solute from snow by meltwaters in advection-dispersion calculations (Hibberd, 1984). Recent models include metamorphism, preferential flow paths, and the solution of ions in snow, and are being used to explore the spatial and temporal distributions of water and solute flux (Iida *et al.*, 2000; Stagnitti *et al.*, 1999). Results suggest that solute should be partitioned into mobile and immobile fractions, whose interaction controls solute release into the meltwater flow (Feng *et al.*, 2001).

Snowmelt-particulate Interactions

Chemical reactions between meltwater and inorganic/organic particles can affect the concentration of solute in meltwater. Many studies have observed the neutralization of snow acidity by carbonaceous dusts from a variety of sources of either local (Colin *et al.*, 1987) or remote origin (Loye-Pilot *et al.*, 1986). Delmas *et al.* (1996) determined that the rate of chemical weathering of dusts in meltwaters depended on the location of the dust in the snow cover; dust in the lower strata of snow showed the highest rates of weathering due to increased partial pressures of CO_2 that arise during dust-meltwater interaction (see Figure 6).

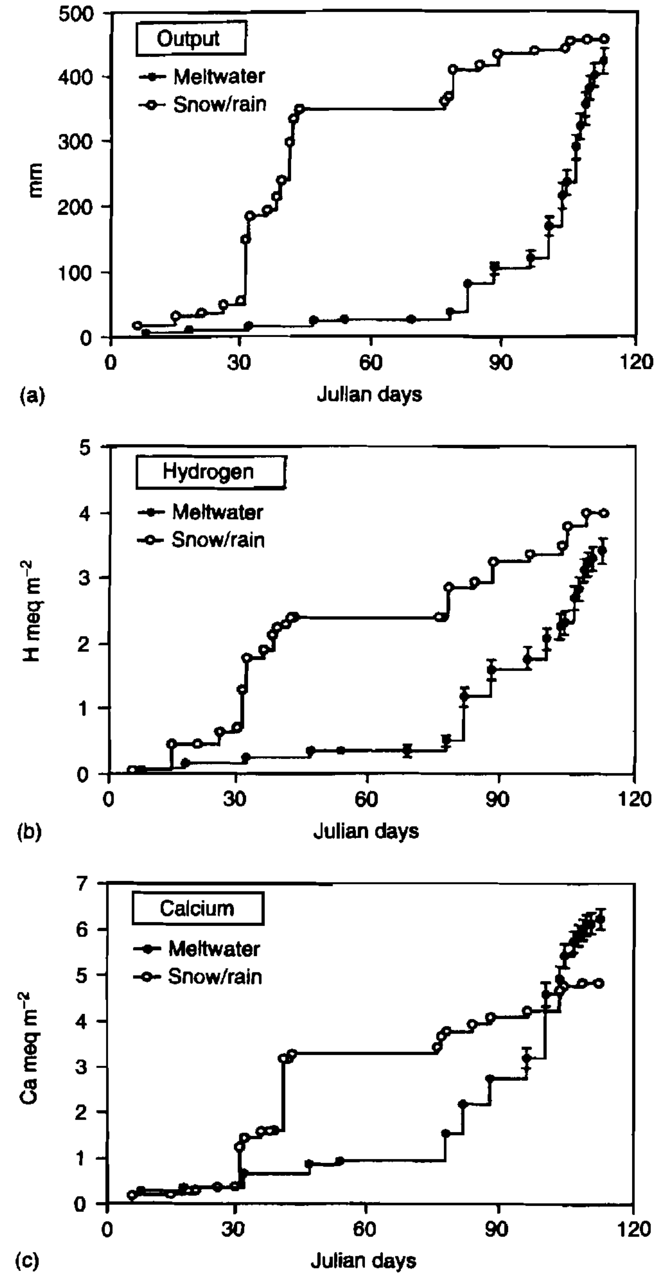

Figure 6 The neutralization of meltwater by calcareous dust in the French Alps. (a) The cumulative input of snow water equivalents and rain to the snowcover, and the cumulative output of meltwater. The water balance is approximately equal. (b) The cumulative input of H^+ to the snow cover, versus the cumulative output. There is a net loss of H^+ during the thaw. (c) The cumulative input of Ca^{2+} to the snow cover, versus the cumulative output. There is a net increase in Ca^{2+} during the thaw (After Delmas *et al.*, 1996)

These lower strata dust particles were, thus, the most efficient at neutralizing acidic meltwaters.

The leaching of litter in snow by meltwaters removes soluble organics and other chemical species (Jones and Sochanska, 1985; Stottlemyer, 1987). Surficial ionic exchange may also take place between meltwaters and the organic debris (Cronan and Reiner, 1983). Leaching experiments (Courchesne and Hendershot, 1988) show that large amounts of PO_4, K, Mn, Ca, and Mg are discharged from litter-laden snow covers, and a decrease in the acidity of meltwaters may arise from cation exchange.

Microbial Activity

During spring melt, the presence of liquid water and the increase in solar radiation stimulate microbiological and invertebrate activity in snow cover. Jones and Debois (1987) showed that meltwater production increased microbiological activity on canopy fallout in forest snowcover. The presence of meltwater also results in photosynthetic activity of truly motile algal populations within the snow cover (Hoham, 1987). Photosynthesis results in an increase in algal biomass at the expense of nutrient concentrations in the meltwaters. Decreases in the concentrations of NH_4 and NO_3 are particularly noted during the growth of algal populations, and may be of the order of 0.67 eq[N] ha^{-1} day^{-1} and 1.05 eq[N] ha^{-1} day^{-1} respectively (Jones, 1999). The loss of nutrients in snow meltwaters over the whole melt season may be appreciable, approaching 20–30% in some years (Jones, 1991).

Snow Nutrient Fluxes and Basin Budgets

Direct acidification from snowmelt remains a concern in many catchments with poor buffering capacity and proximity to certain industrial sources (Tranter *et al.*, 1988a). It is now realized, however, that many episodic stream and lake acidifications that had been associated with the melt of seasonal snowcover were, in fact, due to the mobilization of soil water that carried high ionic loads during the snowmelt period (e.g. Peters and Driscoll, 1987). The role of low ionic strength snowmelt waters is apparently to mobilize geochemical transport from soils in the basin (Hendershot *et al.*, 1992). The exception is where saturated frozen ground with macropores permits runoff with minimal soil interactions (e.g. Jones and Pomeroy, 2001; Quinton and Pomeroy, 2005). The direct delivery of nutrients from snow is now recognized as the primary snow-derived geochemical impact on basin hydro-ecology (Tranter and Jones, 2001). In basins that sustain a long snow-covered period, the major input of N and S to soils and water bodies arises in snow meltwaters in spring. The major output may also occur during the same period when export from the basin by streams is the greatest because of the meltwater runoff (Brooks and Williams, 1999; Brooks *et al.*, 1999). Where basins are poorly buffered, there is a dramatic drop in pH during snowmelt because of acids released directly from snow, and/or mobilized from soil. In either case, runoff during snowmelt has been linked to severe stream and

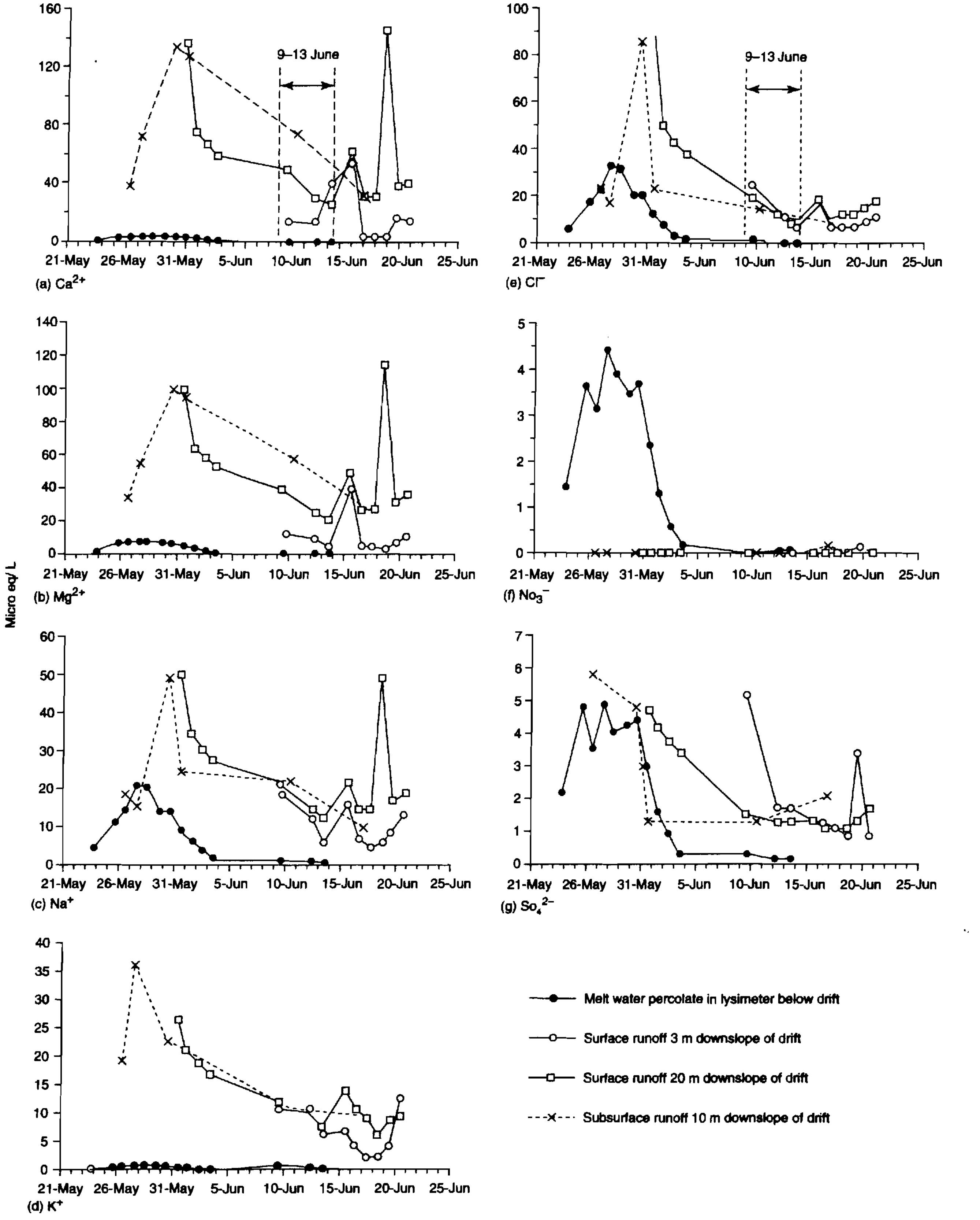

Figure 7 Ion concentrations in (i) meltwater, (ii) surface runoff 3 m downslope, (iii) surface runoff 20 m downslope, and (iv) subsurface runoff through 10 m of peat (After Quinton and Pomeroy, 2005)

lake acidification (Galloway *et al.*, 1987) (*see* **Chapter 95, Acidic Deposition: Sources and Effects, Volume 3**).

On an annual basis, the input and/or output of inorganic nutrients are relatively small, relative to the amount of nutrient that is being recycled within the basin itself. For example, values for the export of NO_3 by streamwater during melt in a boreal forest range from $0.65\,\mathrm{kg\,N\,ha^{-1}}$ to $1.7\,\mathrm{kg\,N\,ha^{-1}}$ (Tranter and Jones, 2001), which is small in comparison to organic pools of N. However, inorganic N has an extremely important role in primary productivity and is linked directly to carbon uptake by ecosystems. The contribution of inorganic N during snowmelt can be one of the largest inputs of this nutrient during the year (Jones, 1991)

The export of N as NO_3 originates both from the solute in the meltwaters (Williams *et al.*, 1993), and/or from the leaching of the species from soil after over-wintering nitrification of organic matter (Peters and Driscoll, 1987; Rascher *et al.*, 1987). In a study of a Colorado watershed, Lewis and Grant (1980) found that hydrological export of N as NO_3 increased significantly after winters over which the soil had frozen. This is supported by the work of Groffman *et al.* (1999) in a northern hardwood forest. The studies of Stottlemyer and Toczydlowski (1990), Stottlemyer and Toczydlowski (1999) indicated that nitrification occurs throughout the winter in organic soils when frozen soil conditions do not occur. Slow sustained meltwater discharge in winter, moves the mineralized N to lower inorganic horizons, where it cannot be taken up by microbiological activity. Some of the NO_3 can then be removed during the main melt period by macropore flow into surface water channels. Heuer *et al.* (1999) show that N dynamics in high altitude-basins are controlled by soil infiltration and transformation of N, and that soils can act as sources (alpine) or sinks (subalpine) of N during snowmelt.

Brooks *et al.* (1996) estimated that the winter/spring N losses by denitrification (N_2, N_2O) from an alpine basin were equal to the NO_3–N input by snowmelt, and represented 50% of the annual gaseous N loss. In this particular system, the loss of N via runoff was negligible during snowmelt. This latter result is in contrast with the hydrologic losses measured by Williams *et al.* (1993) at another alpine site, by Peters and Driscoll (1987) at a hardwood forest site, and by Jones and Roberge (1992) at a coniferous boreal site. These studies show the differing response of ecosystems to the input of N by snowmelt. The factors controlling the hydrologic export of N as NO_3 remain poorly understood.

An example of the effect of a nutrient-poor basin on the chemistry of snowmelt water, as it follows flow pathways to a stream is given by Quinton and Pomeroy (2005) for a frozen soil tundra site (Figure 7). Over flow pathways of from 3 to 20 m from a snow patch, enrichment of Ca, Mg, K, Cl, and Na was substantial. Concentrations increased from one to two orders of magnitude, indicating leaching from soil particles and surface vegetation, and possibly mixing with and flushing of soil water. SO_4 concentrations in meltwater were not strongly modified by hillslope flow, whilst NO_3 concentrations dropped to below detection limits shortly after exposure to the soil, indicating rapid microbiological uptake in the near-surface soil zones, despite below 0 °C soil temperatures.

Summary

Studies of the chemistry of snow-covered basins show that snow is not a passive reservoir of chemical species. Snow-atmosphere exchange by wind redistribution, dry deposition, and volatilization accompanied by various physical processes such as air and water movement, and snow metamorphism within the pack can increase or decrease the quantity of certain species, and/or lead to a redistribution of species within the snow cover. Chemical reactions such as photolysis also take place, while the presence of microorganisms in the snowpack will influence nutrient concentrations during the melt period. Inputs of inorganic nutrients to snow can be an important part of the annual cycling in the basin, particularly for N. Snowmelt chemistry alone is rarely directly responsible for major chemical fluctuations in water bodies, but the meltwater flux is important in mobilizing soil constituents and relocating soil water to the stream.

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