

Snow Ecology

An Interdisciplinary Examination of
Snow-Covered Ecosystems

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3 The Chemistry of Snow: Processes and Nutrient Cycling

MARTYN TRANTER AND H. GERALD JONES

3.1 Introduction: Snow Chemistry and Ecology

The previous chapter demonstrated that the physical characteristics of snow are determining factors in the transfer of energy in snow-covered systems (Pomeroy and Brun, Chapter 2). In this chapter, we demonstrate how the chemical composition of snow plays an important role in biogeochemical cycles, particularly in the supply of nutrients to terrestrial and aquatic ecosystems, because where nutrient reserves are limited, biological productivity may depend heavily on snow meltwater inputs to the soil. In other cases, seasonal snowmelt is a major input to the hydrology of catchments. Water quality and aquatic productivity during meltwater runoff can also depend to a large extent on the chemical composition of the meltwaters.

The chemistry of snow has been studied for two main reasons. The first is because of the potential impact of snow meltwaters on the quality of surface waters (Bales, Davis, and Williams, 1993; Peters and Driscoll, 1987; Schöndorf and Hermann, 1987; Tranter et al., 1988a) and the second is to interpret the climatic and pollution records recorded in cold, dry snow cover (Bales and Wolff, 1995). Acidity in snows derived from the scavenging of pollutants (SO_2 and NO_x) from air masses originating in industrial urban areas (Heubert et al., 1983; Barrie and Vet, 1984; Dasch and Cadle, 1985; Delmas, Briat, and Legrand, 1982; DeWalle et al., 1983) is flushed out of the snowpack early in the melt season, giving rise to fluxes of low pH meltwaters to soils and streams (Hendershot et al., 1992; Goodison, Louie, and Metcalfe, 1986; Galloway et al., 1987). Decreases in aquatic productivity and fish kill during the spring freshet occur (Hagen and Langeland, 1973; Muniz, 1991). Earlier work concentrated on the flushing or elution of solute from the melting snow cover. More recent work has concentrated on other chemical processes that occur in snow cover. It has become clear that certain chemical species can be gained or lost during physical changes in the snow cover (Gregor, 1991), can be transformed under certain meteorological conditions (Pomeroy, Davies, and Tranter, 1991), and can be affected by microbiological activity (Jones, 1991). During

the 1990s, the synergy between snow chemistry and snow ecology was established (Jones, 1999).

Two main types of processes influence the chemical composition of snow and meltwaters: those involving the heat and mass fluxes that occur during sublimation and melting (Davis, 1991) and those involved with chemical transformations (Tranter, 1991). Chemical migration may take place during the former processes, but, in general, the chemical species are conservative in the sense that no chemical transformation takes place. The physical properties of individual species (e.g., solubility, vapour pressure) play a role in determining any change in the chemical loading of the snow cover (Brimblecombe and Shooter, 1991). Chemical transformations may occur by chemical reactions (e.g., oxidation; Bales et al., 1987) or may arise from microbiological activity (e.g., algal assimilation of nitrogen [N] species; Hoham et al., 1989).

Thus, it has become apparent during the past decade that snow is not a passive reservoir of chemical species. It has also become clear that one cannot study the chemical dynamics of snow without taking into account the physical and biogeochemical characteristics of the snow cover environment. This chapter deals with these aspects of snow and the environment through a discussion of the chemical evolution of snow from the time of formation in the atmosphere to the final runoff of meltwaters to stream channels during the ablation season.

3.2 The Chemistry of Snowfall

3.2.1 Formation of Snow Crystals and the Scavenging of Atmospheric Species

The atmosphere and the surface of the biosphere form a complex recycling system that maintains the composition of the earth's gaseous envelope (Mooney, Vitousek, and Matson, 1987). The atmosphere is a dynamic reservoir where terrestrial and marine emissions from the earth's surface are subjected to oxidation processes before being recycled back to the oceans and continental land masses. The main transfer mechanisms are by direct (dry) deposition of chemical species to surfaces or by precipitation (snow and rain) scavenging (Barrie, 1991; Cadle, 1991).

Snowfall contains crustal elements, such as calcium and magnesium, from terrigenous dust (Delmas et al., 1996; Frazén et al., 1994; Hinkley, 1994), anthropogenic pollutants (Landsberger et al., 1989), weak organic acids (Maupetit and Delmas, 1994), neutral organics from natural sources (Likens, Edgerton, and Galloway, 1983), and trace metals (Thornton and Eisenreich, 1982). Snow formation and snowfall incorporate

chemical species from the atmosphere by three main processes: imprisonment during initial formation of ice crystals; capture of gases, aerosol, and larger particulates within clouds; and scavenging of these materials below the cloud layers during snowfall (Barrie, 1991).

Cloud water droplets contain solute as a result of aerosol scavenging and diffusion of atmospheric gases into solution. The soluble species are mainly NH_4^+ , SO_4^{2-} , NO_3^- , Ca^{2+} , K^+ , and Mg^{2+} , derived from natural and anthropogenic emissions, in addition to Na^+ and Cl^- from sea-salt aerosol. Ice crystals initially form in the atmosphere by the freezing of supercooled cloud water droplets at temperatures $> -40^\circ\text{C}$ (see Figure 3.1), which is catalysed by ice nuclei that usually consist of sea-salt aerosols, particulate organic debris, and/or fine particulate clays (Kamai, 1976; Pomeroy and Brun, Chapter 2). Ice crystals may form by direct deposition of water vapour onto ice nuclei and by water droplets' either touching ice nuclei (contact nucleation) or scavenging ice nuclei, so that ice crystals form within the water droplet (immersion-freezing nucleation). Solute does not fit into the ice crystal lattice particularly well (Colbeck, 1981). Hence, most solute is rejected into the outer edges of the ice crystal during freezing.

After nucleation, the ice crystals may grow by direct deposition of water vapour onto the ice crystal surface at the expense of supercooled water droplets in the immediate vicinity. Dendritic, needle and plate-like crystals are the main result of this process (Magono and Lee, 1966). These growing ice crystals contain very low solute concentrations (Scott, 1981; Hewitt and Cragin, 1994), largely limited to the original, imprisoned nuclei. Limited amounts of compounds with relatively high vapour pressure – e.g., HNO_3 , HCl – may be incorporated into the growing crystal by absorption or adsorption. In-cloud conditions favouring collisions between growing crystals and supercooled droplets result in the formation of rounded crystals known as rimed snow or soft snow pellets (graupel). These types of crystals contain much larger amounts of solute than crystals formed mainly by vapour transport (Cerling and Alexander, 1987), because much of the solute contained in the supercooled droplets is captured by rapid freezing. For example, at Dye 3 in Greenland, rimed snow crystals compose ~ 5 percent of the snowpack and contain ~ 30 percent of the solute (Borys et al., 1993).

Further scavenging of aerosols and other particulates occurs below clouds by adsorption or impaction onto the surface of all types of crystals during snowfall (Scott, 1981). Snow is a better scavenger of particulate material than rain (Raynor and Haynes, 1983; Leuenberger et al., 1988; Nicholson, Branson, and Giess, 1991). The efficiency of scavenging is related to the surface area:mass ratios of the crystals as they sweep out

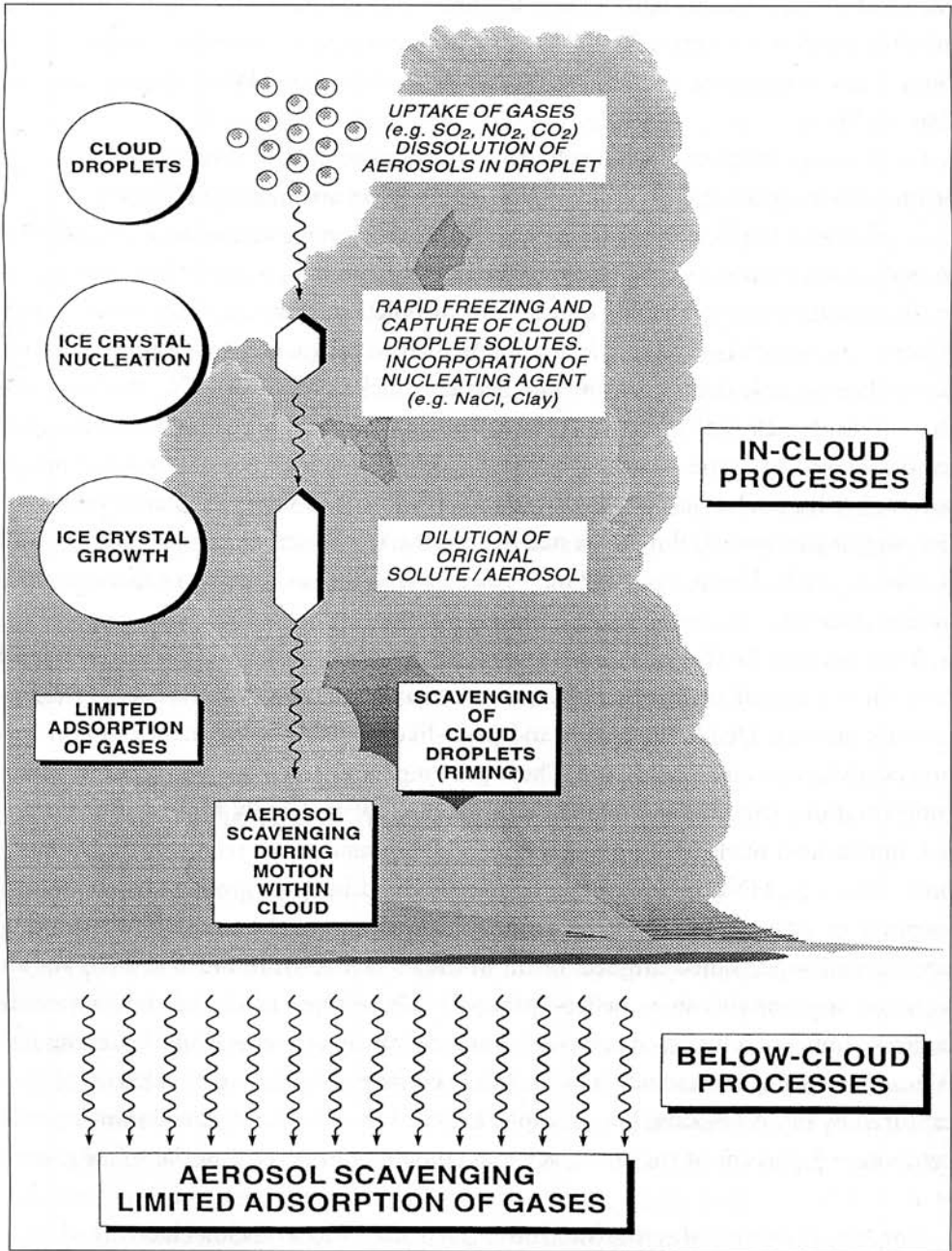


Figure 3.1. The main processes that influence the chemical composition of snow crystals during growth and fall.

the air during fall. The overall scavenging efficiency of precipitation can be expressed either as a washout ratio, W , or as a scavenging coefficient, ω (Misra et al., 1985). The values of these parameters for removal of chemical species from the atmosphere by precipitation are not constant and depend on meteorological conditions and the type of precipitation. The washout ratio is dimensionless and is expressed as the ratio of the amount of a chemical species in precipitation to that in the air,

$$W = \rho_a(C_p/C_a) \quad (3.1)$$

where ρ_a is the density of air (g m^{-3}), C_p is the concentration of the species of interest in precipitation ($\mu\text{g g}^{-1}$), and C_a is the concentration of that species in air ($\mu\text{g m}^{-3}$).

The scavenging coefficient ω (s^{-1}) can be thought of as a rate constant for the transfer of species from air to precipitation on an event basis,

$$\omega = \ln[C_t/C_0]/t \quad (3.2)$$

where C_t is the concentration of the species of interest ($\mu\text{g m}^{-3}$) in air at time t (s), and C_0 ($\mu\text{g m}^{-3}$) is the concentration of the species in air at time zero (Raynor and Haynes, 1983).

Table 3.1 shows typical values for W and ω for some of the more common chemical species found in snow.

General relationships between the formation of snow crystals (based on snow crystal type) and the amount of scavenged solute have been found, despite the complexity of physical and chemical processes outlined above, the large range in the concentration of chemical species, and the variable values found for W and ω . Lamb, Mitchell, and Blumernstein (1986) found that the highest concentrations of solute were associated with the smallest unrimed crystals, demonstrating the importance of surface area: mass ratios as a scavenging parameter. Borys et al. (1983) showed that riming was also a factor in the ability of snowfall to scavenge the atmosphere as, in general, the solute content of snow crystals increased with the degree of riming. Hewitt and Cragin (1994) isolated individual snowfall crystals by crystal habit and 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. The reason for this phenomenon is unknown. The distribution of certain species within, or on, different individual crystals in snowfall has been reported (Kamai, 1976). Rango, Wergin, and Erbe (1996) further suggested that the preferential incorporation of rime on certain snowfall crystal features, such as edges, should be reflected by pollutant distribution at the scale of individual crystals.

Table 3.1. Removal of chemical species from the atmosphere by snow: washout ratios (W ; dimensionless) and scavenging coefficients (ω , s^{-1}).

Species	Washout ratio (W)		Reference	Scavenging coefficient (ω , s^{-1})	
	Mean*	Range†			Reference
NH_4^+	410	40–2,200	1		
SO_4^{2-}	570	20–3,000	1, 2, 3, 4	5×10^{-5}	3
NO_3^-	1,000		1, 3	5×10^{-5}	3
SO_2	200	20–500	1		
HNO_3	3,600–5,000‡		1, 3	25×10^{-5}	3
Ca	2,600	170–8,190	1		
K	450	80–4,600	1		
Cr	530	200–1,280	1		
Cu	1,220	60–3,800	1		
Fe	1,160	10–7,110	1		
Ni	1,190	280–3,350	1		
Pb	240	20–1,350	1		
Zn	2,470	280–1,1500	1		

*Mean values from reference 1.

†Range of all references cited.

‡Means from references 1 and 3.

References: 1, Cadle, Dasch, and Mulawa (1985); 2, Scott (1981); 3, Heubert et al. (1983); 4, Davidson et al. (1987).

3.2.2 Spatial and Temporal Variability of Snowfall Composition

The chemical composition of snowfall depends on factors such as the origin of the air masses that are scavenged, the altitude at which snow is deposited, and the meteorological conditions during snowfall (Colin et al., 1989; Davies et al., 1992). Maritime air masses give rise to snow containing mostly Na^+ and Cl^- (Tranter et al., 1986), whereas polluted air masses from industrial areas deposit snow that is highly acidic because of the presence of strong acid anions (NO_3^- , SO_4^{2-}) from fossil fuel combustion (Davies et al., 1984; Landsberger et al., 1989). Table 3.2 shows the chemical concentrations of snowfalls in different regions of the world.

The chemical composition of snow may show spatial variability on length scales of metres through tens of kilometres (Tranter et al., 1987; Pomeroy, Marsh, and Lesack, 1993; Suzuki, 1987; Laird, Taylor, and Kennedy, 1986), reflecting factors such as the proximity to pollution sources, the impact of wind redistribution (Pomeroy and

Table 3.2. Chemical composition of snowfall at selected sites throughout the world.

Location	pH	Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	NH ₄ ⁺	NO ₃ ⁻	SO ₄ ²⁻	Cl ⁻	Source
European Alps	4.4-5.3	18-49	3-15	3-27	1-6	17-60	12-46	28-68	8-32	Puxbaum, Kovar, and Kalina (1991)
Central Asian Mountains	*	19-70	*	1-44	*	*	2.9-60	2.2-51	1-32	Lyons, Wake, and Mayewski (1991)
Turkey Lakes Watershed, SE Canada	4.57	34	0.9	10	0.2	7.5	19	17	3.7	Semkin and Jeffries (1988)
Mid-Wales	3.9-4.5	4-14	4-11	13-30	1-5	*	11-64	16-78	21-69	Reynolds (1983)
Sapporo, Japan	4.4-6.4	13-63	18-67	59-190	2.3-6.4	*	*	70-99	63-310	Suzuki (1987)
Cairngorms, Scotland	4.4	2.5	11	52	2.1	9.8	20	26	91	Davies et al. (1992)
Svalbard	5.4-6.7	0-46	0-200	4-2000	0-96	*	0-7	0-240	0-2400	Hodgkins, Tranter, and Dowdeswell (1997)
South Pole	5.4	*	0.16	0.63	0.03	0.16	1.4	1.5	1.3	Legrand and Delmas (1984)

*Missing values. Single values refer to volume-weighted mean concentrations. All units (except pH) are $\mu\text{Eq/L}$.

Brun, Chapter 2), and differential scavenging by vegetation. Snowfall at high altitudes usually contains lower concentrations of chemical species than at lower altitudes because the depth of air column available for aerosol scavenging is smaller. In addition, air masses subjected to orographic rise release relatively concentrated deposition at lower altitudes and relatively clean snow at higher altitudes (Laird et al., 1986; Lyons, Wake, and Mayewski, 1991). However, in the case of some chemical species (e.g., volatile organochlorines), the concentrations in snow can increase with altitude. This is due to volatilisation of the compounds from relatively "warm" snow at low altitudes and subsequent "cold condensation" in snow precipitated at cooler higher altitudes (Blais et al., 1998). Meteorological conditions during snowfall also influence the chemical composition of snow crystals. Pomeroy et al. (1991) demonstrated that the solute content may change during windblown snow events, related to the extent of sublimation and aerosol scavenging.

The temporal variability of the chemistry of individual snowfalls (Colin et al., 1987) is due to the progressive scavenging of the chemical species during the snowfall event (Raynor and Haynes, 1983). Chemical concentrations often decrease exponentially with time. This gives rise to the use of the scavenging coefficient, ω , to characterise the efficiency of precipitation in removing pollutants from the atmosphere. Recently, transfer functions have been used to characterise the relationship between concentrations of chemical species in the atmosphere and those found in snow (Bales and Wolff, 1995). These functions are complex. They account for not only the removal of species from the atmosphere but also for postdepositional changes in cold snow covers. The latter depend on both the reversible physical exchange of species with the atmosphere and chemical reactions at the snow surface and within the snow cover.

3.3 Chemistry of Cold, Dry Snow Cover

Snow on the ground becomes consolidated into snow cover during cold ($< 0^{\circ}\text{C}$) weather. The temporal and spatial variation in the chemical composition of snowfall usually produces a snow cover that is chemically heterogeneous. Cold snow covers are subjected to a number of physical (Pomeroy and Brun, Chapter 2) and chemical processes (Jones and Stein, 1990; Bales, 1991), which further modify the chemical concentrations of individual snow strata and the total chemical load of the snow cover. The main processes (see Figure 3.2) are surface exchange at the snow-atmosphere interface (dry deposition and volatilisation), surface and subsurface chemical reactions, snow-grain metamorphism within the pack and, in the case of

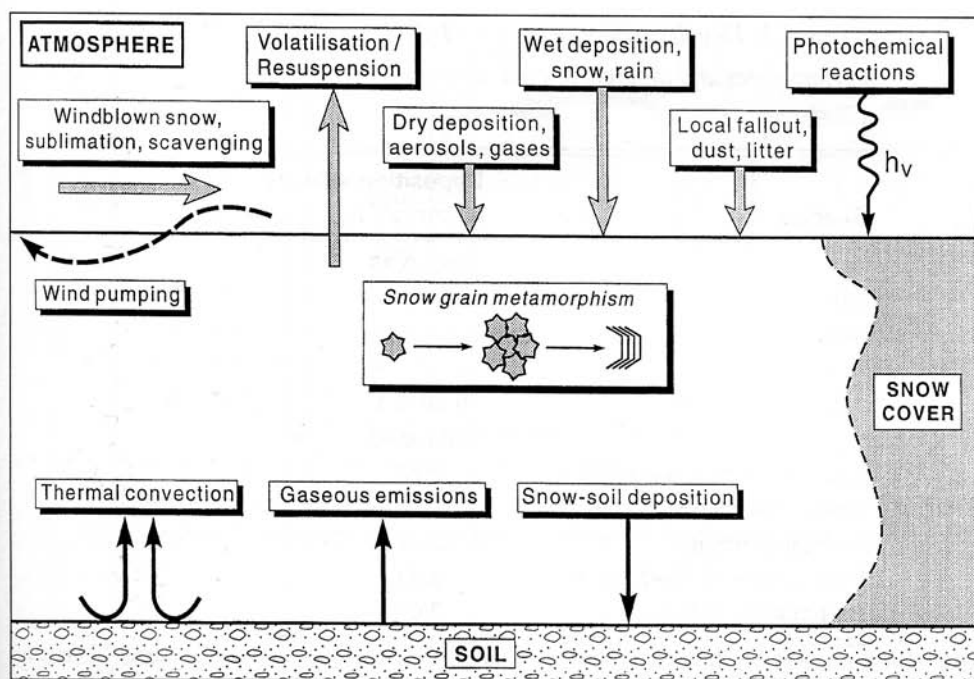


Figure 3.2. The main physical and chemical processes that influence the chemical composition of cold, dry snow cover during the accumulation season.

seasonal ground cover, basal-exchange processes at the snow–soil interface (e.g., gaseous emissions from soil).

3.3.1 Processes at the Atmosphere–Snow Surface Interface

3.3.1.1 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, whereas gaseous species may also be adsorbed (Conklin, 1991). In general, dry deposition to snow cover is much lower than to most other snow- and ice-free terrestrial surfaces, because the surface roughness of snow is much lower (0.01 cm compared with 0.03–100 cm; Cadle, 1991). Hence, the relative magnitude of dry to wet deposition is smaller for snow- and ice-covered terrain than for other terrestrial surfaces.

The net dry deposition flux (F_x , $\mu\text{eq m}^{-2}$) of a species (X) is expressed as the product of the atmospheric concentration (C_x , $\mu\text{eq m}^{-3}$) of the species and the deposition velocity ($V_{d(x)}$, m s^{-1}), so that

$$F_x = C_x V_{d(x)} \quad (3.3)$$

Table 3.3. *Dry deposition velocities (V_d ; cm s^{-1}) for various gases and aerosols to snow surfaces (after Cadle, 1991).*

Species	Deposition velocity V_d (cm s^{-1})
SO ₂	0.02–0.25
NO	<0.03
NO ₂	0.005–0.12
HNO ₃	<0.02–2.0
NH ₄ ⁺	0.08–0.1
O ₃	0.01–0.08
Organic carbon aerosols	0.28
Elemental carbon	<0.2–0.55
Crustal aerosols	0.2–3.4
Particulates (0.15–0.3 μm)	0.03
Particulates (0.5–1 μm)	0.02

F_x is negative when there is net deposition to the snow surface. The deposition velocity, $V_{d(x)}$, is usually specified with reference to a height above the snow surface (e.g., 1–2 m), because the flux, rather than the atmospheric concentration, C_x , is independent of height. Deposition velocities vary with meteorological conditions, physical characteristics of the snow surface, and the physical properties and chemical reactivity of the species. Bales et al. (1987) found that dry deposition rates to new snow were higher than those to old snow. This was believed to be the 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, Barrie, and Fanaki (1983), who suggested that the interception of aerosols by ice needles in relatively fresh snow contributed significantly to the measured rates of dry deposition.

Cadle, Dasch, and Mulawa (1985) calculated that V_d for HNO₃ was approximately one order of magnitude greater than for SO₂. The difference in deposition rates 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 SO₂ to SO₄²⁻ at the air–crystal interface (Bales et al., 1987; Bales, 1991). The rate of deposition of HNO₃ is much higher to wet snow than to cold, dry snow (Cadle, 1991). Values of V_d for various chemical species (gases and aerosols) to snow are shown in Table 3.3. The values are within the range $0.02\text{--}3 \times 10^{-2} \text{ m s}^{-1}$. At these levels of deposition velocity, dry deposition can be a significant factor in the

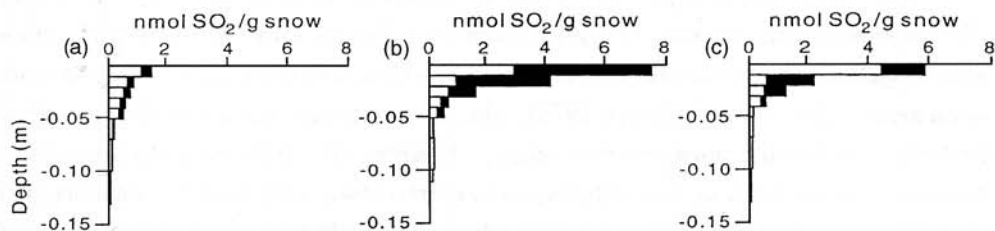


Figure 3.3. Impact of dry deposition on the SO_2 concentration of surface snow (after Valdez et al., 1987). Black bars show increases in SIV (SO_2) concentrations over a 6-hour period, white bars show increases in SVI (HSO_3^- ; SO_4^{2-}) concentrations. Conditions are: (a) Temperature, -2.2°C ; SO_2 mixing ratio, 81 ppbv; age snow, 112 days; density, ρ_s , 400 kg m^{-3} . Measured dry deposition velocity, $2.0 \times 10^{-4} \text{ m s}^{-1}$. (b) Temperature, 0°C ; SO_2 mixing ratio, 70 ppbv; age snow, 9 days; density, ρ_s , 380 kg m^{-3} . Measured dry deposition velocity, $9.3 \times 10^{-4} \text{ m s}^{-1}$. (c) Temperature, 0°C ; SO_2 mixing ratio, 79 ppbv; age snow, 10 days; density, ρ_s , 400 kg m^{-3} . Measured dry deposition velocity, $5.9 \times 10^{-4} \text{ m s}^{-1}$.

chemical composition of snow. For example, a fresh snowfall of 10 mm snow water equivalent (SWE) containing $600 \mu\text{g L}^{-1}$ of HNO_3 (i.e. a load of 6 mg m^{-2}) will double the load of HNO_3 (to 12 mg m^{-2}) within 2 days of being in contact with an air mass containing an average HNO_3 concentration of $2.5 \mu\text{g m}^{-3}$. Field studies in North America suggest that the overall contribution of dry deposition to the chemical composition of snow in the case of SO_4^{2-} and NO_3^- is approximately 20–25 percent (Cadle and Dasch, 1987; Barrie and Vet, 1984; Cadle, 1991). Figure 3.3 shows an examples of the impact of dry deposition on SO_2 concentrations in surface snow, which depend on snow temperature ($^\circ\text{C}$), age (days) and density of snow (ρ_s), and the mixing ratio of SO_2 in air (ppbv). (Valdez et al., 1987).

Snow covers under forest canopies may receive greater contributions of dry deposition. Dry deposition velocities of gases and aerosols to vegetative surfaces are higher than those to snow (Höfken, Meixner, and Ehhalt, 1983; Dasch, 1987), so the removal of adsorbed species from the canopies by throughfall, wet snow, and/or rain can contribute significantly to the chemical load of the snow cover. Aeolian dust and other particulate matter are continuously being deposited on snow covers. The result of dust deposition usually is to reduce the acidity of snows (Sequeira, 1991), particularly during melt periods (Clow and Ingersoll, 1994; Delmas et al., 1996).

Throughout the winter, cold snow also accumulates nutrients by the deposition of biological debris either as fallout from above the snow surface or by incorporation within the snow cover itself. In forest snow covers much of the deposition arises

as litterfall from the canopy (Jones and Debois, 1987) and mammalian excrement (White, Garroff, and Heisey, 1997). Invertebrate fallout from wind-borne arthropods and winged invertebrates may also contribute nutrients to snow, particularly in more open areas (Edwards and Banko, 1976), whereas vertebrate and invertebrate activity under and within the snow cover (Aitchison, Chapter 5) transforms and redistributes nutrients from the soil upward into the snow matrix. Inorganic N and phosphorus [P] species are readily bioavailable for snow algae, which develop in the latter stages of the spring melt (Hoham, 1987). Organic material, particularly excrement, serves as a substrate for heterotrophic organisms such as fungi (Stein and Amundsen, 1967). Biological deposition thus may be an important component of nival food webs (Hoham and Duval, Chapter 4; Aitchison, Chapter 5).

However, the amount of nutrients deposited by animals and plants on snow that is available to organisms is difficult to evaluate. This is due to the extremely high spatial variability of the sources. Jones (1991) attempted to calculate nutrient inputs to snow from different vertebrates by considering population densities and the amount and chemical composition of animal excrement. In the case of animals that herd (e.g., deer), local (5- to 10-ha scale) deposition rates may be relatively high ($0.2 \text{ kg N ha}^{-1} \text{ day}^{-1}$). On the same scale, solitary mammals, such as moose and hares, deposit only 1.5×10^{-4} to $2.5 \times 10^{-3} \text{ kg N ha}^{-1} \text{ day}^{-1}$. Although not strictly applicable to the estimations of nutrient contents of cold snow cover, data published by Edwards and Banko (1976) indicate that the pool of particulate nutrient (debris, dead and living invertebrates) mostly blown onto snowpatches from the surrounding cover contains up to $0.045 \text{ kg N ha}^{-1}$.

3.3.1.2 *Volatilisation*

In general, the dry deposition of aerosol species to snow is considered to be irreversible (Bales and Wolff, 1995). However, recent work has suggested that there may be a concomitant loss of some ionic species (in particular, SO_4^{2-}) during sublimation of snow crystals, but there is no known mechanism to explain such a loss (Cragin and McGilvary, 1995). Laird, Buttry, and Sommerfeld (1999) found no loss of HNO_3 adsorbed on artificial snow in columns sealed from ambient air. However, they obtained evidence that a small amount (≤ 10 percent) of HNO_3 can exist as vapor within the interstitial space of the snow matrix under certain conditions because of volatilisation of HNO_3 from the ice surfaces. In open exposed natural sites, wind transport has the potential to change the chemical composition of snow due to three main physical processes: namely sublimation of water vapour, scavenging of

aerosols and gases from the atmosphere, and volatilisation (Pomeroy and Jones, 1996). Pomeroy et al. (1991) found evidence that both gains (scavenging) and losses (volatilisation) of NO_3^- and NH_4^+ could take place at different stages during windblown snow events.

Some chemical species, particularly organic species, may volatilise directly from snow to the atmosphere because of their relatively high vapour pressures. For example, Hogan and Leggett (1995) showed that synthetic organic compounds (paranitrotoluene, dinitrotoluene, and nitroglycerine) stored in cold snow cover are lost as the snowpack ripens. Organochlorines (e.g., hexachlorobenzene) may be lost from arctic snow cover by volatilisation during the summer (Gregor, 1991).

3.3.1.3 *Photochemical Reactions in Surface Snow Cover*

Snow is in intimate contact with the atmosphere, and oxidation of certain species by atmospheric oxidants may take place on cold snow grain surfaces, particularly 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), $\text{SO}_3^{2-}/\text{SO}_4^{2-}$, based on the known oxidation rates by H_2O_2 , O_3 , and O_2 . In addition to the presence of liquid films, however, light may also play a key role in the chemistry of snow cover by initiating photochemical reactions.

It is known that ice crystals play an active role in the photochemistry of the upper atmosphere (Molina et al., 1987) and of clouds (Mitra, Barth, and Pruppacher, 1990). Ice crystals on the ground thus have the potential for photochemical reactions. Photoreactive species found commonly in snow include peroxides (Nefstel, 1991), trace metals (Rahn and McCaffrey, 1979), and NO_3^- . Although unequivocal evidence of photochemical processes in snow has yet to be obtained, some workers have reported changes in the chemical composition of snow that are consistent with photochemical reactions.

Palmer, Smith, and Neirink (1975) reported the presence of O_3 in the surface snows of a high mountain valley, which suggests that photooxidation reactions may have taken place involving species such as the OH radical, H_2O_2 , O_3 , and organic peroxides (Gunz and Hoffmann, 1990a, 1990b). Sigg, Nefstel, and Zircher (1987) measured the decrease in concentrations of H_2O_2 in surface alpine snows and proposed that photolysis was the primary mechanism. Photochemical reactions in snow are complex and are difficult to distinguish from other processes (e.g., dry deposition) related to exchange at the snow-atmosphere interface. 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 evaporation of HNO_3 from snow during metamorphism, but they were unable to distinguish between the two mechanisms. Evidence for the photochemical production of NO and NO_2 (NO_x) in Antarctic snow cover has been recorded by Jones et al. (2000). However, they were not able to demonstrate that NO_3^- was the precursor of NO_x . The duration of exposure and the intensity and wavelength of light are principal factors in photochemical reactions. For example, Duchesneau (1993) found that the apparent dry deposition rate of NO_3^- to snow cover on a high-elevation ice cap differed in an area exposed to radiation from that in an area deliberately shaded by a flysheet. It was suggested that the concomitant photodegradation of HNO_3 during the deposition process was responsible for the lower dry deposition rate in the irradiated area. However, a similar experiment over a shorter time period in a subarctic site gave no such result (Jones et al., 1993), and it is believed that both the exposure time and the higher degree of light intensity and ultraviolet radiation at the high-elevation site were responsible for the difference between the two snow covers.

In addition to organic peroxides, other photoreactive organic species are found in snow. Jaffrezo, Clain, and Maschet (1994) reported that the concentrations of benzo[*a*]pyrene in Greenland snow cover dropped by over 90 percent during post-deposition modification. It was suggested that photochemical degradation of the benzo[*a*]pyrene took place because of the presence of OH radicals from H_2O_2 decomposition.

3.3.2 In-Pack Processes

3.3.2.1 *Metamorphism*

Metamorphism of ice crystals commences almost immediately after deposition (Colbeck, 1987; Davis, 1991). Metamorphism of dry snow crystals occurs because of redistribution of water vapour between and within crystals as water vapour is transferred from surfaces of high curvature to sites of low curvature. The crystals lose their characteristic dendritic, stellar, or needle structure and are reconstituted as spherical snow grains (Pomeroy and Brun, Chapter 2). Solute may also become redistributed and concentrated on the snow grain surfaces or, in the case of grain aggregations, along the snow grain boundaries. Further metamorphic processes follow that lead to growth of larger snow grains and faceted crystals (Colbeck, 1987). Large rounded snow grains grow at the expense of smaller grains when temperature gradients within the snow cover are small. Grains that lose water vapour should become more concentrated in ionic species if these solutes have negligible vapour pressures.

Growth of the larger grains by crystallisation of water vapour on the grain surfaces should lead to more dilute concentrations of solutes. It is not known to what extent this concentration-dilution process occurs in dry snow (Granberg, 1985), as grain clusters, composed of small and large grains physically bound together, are often formed (Colbeck, 1987). In addition, the loss of all, or almost all, the water from a grain would cause this simple concentration-dilution model to break down, as highly concentrated small droplets may be subjected to phoretic transport to grain surfaces. The growth of grains probably is also accompanied by solute exclusion from the growing ice lattice. Thus, the net effect of this type of metamorphism is believed to be a concentration of solute onto or near the surfaces of ice crystals (Bales, 1991; Colbeck, 1981, 1987; Davis, 1991). The solute may be located in a quasi-liquid surface layer that is believed to exist at temperatures $> -35^{\circ}\text{C}$ (Ushakova and Troshkina, 1974), as discrete aerosol or as concentrated, "doped ice" pockets (Davis, 1991).

The growth of large faceted crystals occurs (at the expense of rounded grains) if temperature gradients are large (Colbeck, 1987). Such crystals are known as the kinetic-growth form and are common in depth hoar (Pomeroy and Brun, Chapter 2). Some studies have shown that formation of depth hoar is accompanied by a loss of ions. As ionic species are assumed to be conserved without chemical change in cold dry snow, loss must be 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^{2-} was lost during depth hoar formation. They suggested that a process that could physically transfer small concentrated SO_4^{2-} particulates away from the depth hoar was responsible for the losses. By contrast, Pomeroy et al. (1993) found that SO_4^{2-} and Cl^- concentrations in depth hoar increased in proportion to the overall loss of water vapour from the depth hoar to adjacent snow layers. However, NO_3^- concentrations remained approximately constant, indicating a concomitant loss of the species. The loss was believed to occur via HNO_3 vapour. In summary, the formation of kinetic-growth crystals seems to lead to the loss of some species, but the precise mechanisms are unknown.

3.3.2.2 *Windpumping and Thermal Convection*

Cyclic air movement between the atmosphere and snow cover occurs when wind conditions are favourable (Colbeck, 1989). The phenomenon is known as wind-pumping and theoretically could increase or decrease the concentrations of chemical species in the snow cover, because aerosols and gases could be filtered from the air or removed from interstitial air pockets. Gjessing (1977) simulated natural windpumping

by forcing air through snow and reported that concentrations of K^+ , NO_3^- , and SO_4^{2-} increased in the snow cover.

If temperature gradients increase to a critical threshold level, air movement may be initiated in the snow cover by thermal convection (Pomeroy and Brun, Chapter 2), and convective air currents (Sturm, 1991) could also redistribute chemical species throughout the pack. However, no evidence for such redistribution has been documented.

3.3.3 Basal Gas Exchange Between Snow and Soil

Soil microorganisms produce trace gases by a variety of processes, such as respiration, denitrification, and nitrification (Granli and Bøckman, 1994). The onset of snow cover usually results in a decrease in the overall rate of microbiological activity in the soil due to the decrease in temperature at the snow–soil interface. However, trace gas emissions under snow can still represent a significant fraction of annual emissions (CO_2 , >20 percent [Winston et al., 1995]; N_2O , ~50 percent [Brooks, Williams, and Schmidt, 1996]). The extent to which seasonal snow cover influences trace gas emissions from soil varies with the duration and depth of snow during the cold, dry accumulation period and with meltwater discharge and chemistry during the melt season. Thus, wintertime emissions may vary considerably from year to year (van Bochove et al., 2000). Snow is a porous medium, so trace gases released from the soil permeate the snow cover and give rise to either consistent gaseous concentration profiles (Sommerfeld, Mosier, and Musselman, 1993; Jones, van Bochove, and Bertrand, 1999) or ephemeral localised gas-rich pockets of air within the snow cover (Zimov et al., 1993), depending on soil processes and the physical characteristics and dynamics of the snow (see Figure 3.4).

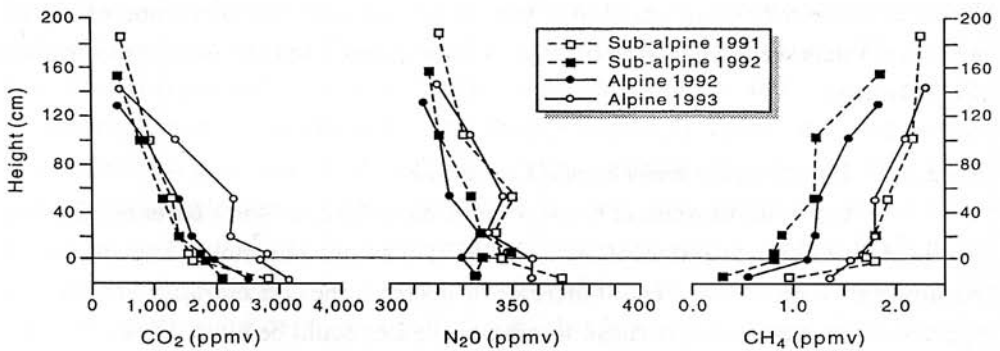


Figure 3.4. Profiles of the gaseous concentration of CO_2 , N_2O , and CH_4 in alpine snow cover (after Sommerfeld, Mosier, and Musselman, 1993).

The soil beneath deep alpine snow cover rarely freezes because of the relatively thick (2–3 m) snow cover. Microbiological activity in the soil is thus maintained throughout the winter (Massman et al., 1995). The continual production of trace gases and a relatively homogeneous snow structure result in consistent concentration profiles of gases with snow depth. Typical profiles are as follows: CO_2 , 1,800–2,500 ppmv m^{-1} (Solomon and Cerling, 1987; Massman et al., 1995); CH_4 , 0.5 ppmv m^{-1} (Sommerfeld et al., 1993); N_2O , 15 ppmv m^{-1} (Sommerfeld et al., 1993). In the case of CH_4 , the flux is from the atmosphere to the soil, showing that the soil was a sink for CH_4 (Figure 3.4). Windpumping can affect the gaseous fluxes through snow by modifying concentration gradients (Massman et al., 1997); the pressure pumping effects are more pronounced when the gradients are weak.

The combination of shallow snow cover and extreme cold air temperatures in arctic and subarctic environments causes the surface soils to freeze, in contrast to deep alpine snows. Microbiological activity ceases when the temperature of the frozen soil drops below -8°C (Coxson and Parkinson, 1987). However, the soil still can remain a source of trace gases even if biological activity in the upper layers of soil becomes dormant. Gases may be released from deeper soil horizons; for example, Zimov et al. (1993) showed that respiration in deep subsurface layers is a source of CO_2 in Siberian soils even in the coldest midwinter period. However, the emissions of gas at the snow–soil interface was episodic and spatially variable, in contrast to the case of deep alpine snow. This is due to the random nature of the physical stresses in frozen soil that mediate the release of the gases to the snow cover. Gases can be released from frozen soils even in the absence of microbiological activity. Coyne and Kelley (1971) believed that the source of CO_2 in snow-covered soil in the Arctic under conditions of extreme cold was due to the release of pockets of CO_2 from within or below the frozen soil layer formed by exclusion of the gas from the soil–water matrix during freezing of the soil in early winter. Thermal stress in mid- and late winter periodically occurred and the resulting episodic and highly variable release of CO_2 was similar to that observed by Zimov et al. (1993).

Finally, pockets of gas liberated from soil may remain trapped locally within the snow structure if the permeability of snow cover in an area is low because of the presence of wind-packed snow or ice layers (Hardy, Davis, and Winston, 1995; Jones et al., 1999). Winston et al. (1995) believed that the low values of CO_2 emissions measured over snow in open areas of a boreal forest site were due to the trapping of the gas under impermeable ice lenses formed during melt-freeze cycles. In open exposed subarctic snow covers, subsurface trapping of gases and windpumping can mediate gaseous fluxes between the soil and the atmosphere (Jones et al., 1999).

3.4 Chemistry of Wet Snow and Snow–Meltwater Systems

The cold ($<0^{\circ}\text{C}$) accumulation winter period is followed by the spring melt in many temperate and higher latitude environments. The physical aspects of meltwater production are covered in Chapter 2 by Pomeroy and Brun. Snow meltwaters penetrate dry snow cover and produce a wetting front that separates an upper layer of wet isothermal snow at 0°C from a lower layer of dry snow with a temperature $<0^{\circ}\text{C}$. Flow fingers develop at the leading edge of wetting fronts due to flow instabilities and structural discontinuities in the snow (crusts and ice lenses). The permeability of the flow fingers increases because of grain growth in wet snow; hence, these flow fingers become areas of higher flow discharge rates called macropores (Kattleman, 1985, 1989; Marsh, 1990, 1991). Rain-on-snow events generally accelerate the growth of macropores.

Percolation of meltwaters through the snow cover 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 various physical and biological processes (see Figure 3.5). These processes are the leaching of solute from

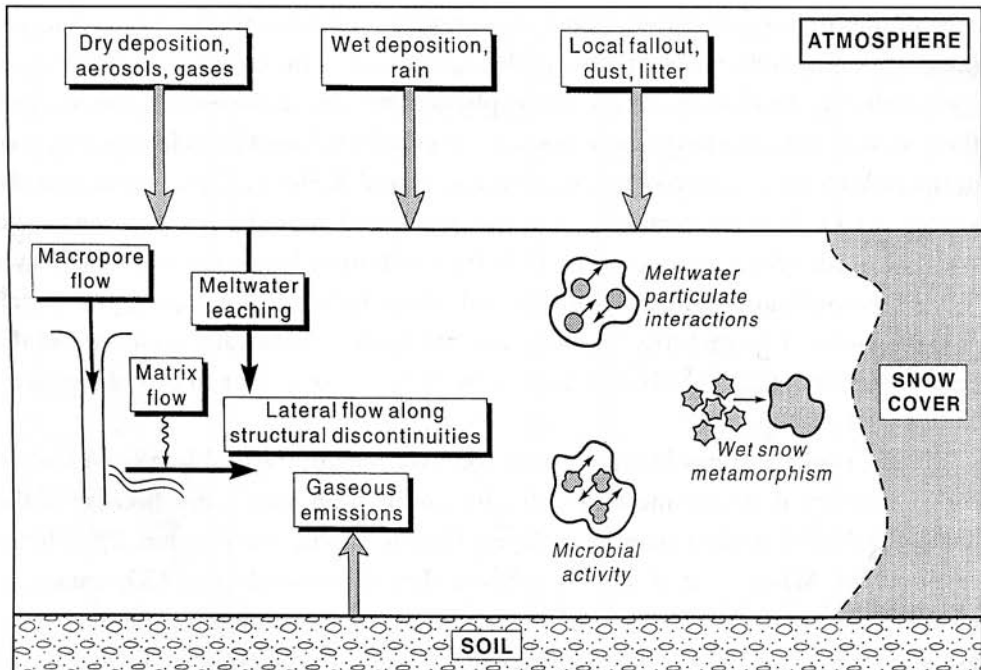


Figure 3.5. The main physical, chemical, and biological processes that influence the chemical composition of snow cover during the thaw.

snow grains and crystals, meltwater–particulate interactions, and 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 will also influence the chemistry of meltwaters (Tranter et al., 1992).

3.4.1 Leaching of Solute from Snow Crystals

The leaching of snow grains by meltwater causes fractionation of solute species between the grains and the liquid medium, and the meltwater front becomes progressively more concentrated as it moves through the pack (Tranter, 1991). The degree of fractionation of any solute species, x , between snow and meltwater is expressed by a nondimensional concentration factor, CF_x ,

$$CF_x = {}^xM_m / {}^xM_p \quad (3.4)$$

where xM_m is the concentration of species x in any meltwater fraction and xM_p is the concentration of x in the parent snow *prior* to melt. Values of CF_x 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_x decreases as melt progresses and the snowpack is leached to values of <0.1 in the final meltwaters. The efficiency of meltwater leaching (i.e., high values of CF in initial meltwater discharges) depends on the micro- and macroscale distribution of solute in snow grains and meltwater hydrology.

The microscale distribution of solute in snowpacks before melt is influenced by snow metamorphism (Davis, 1991; Pomeroy and Brun, Chapter 2). Highly concentrated solute is believed to be present initially as discrete droplets of liquid residing in the boundaries between associated snow grains and as quasi-liquid films on grain surfaces. (Mulvaney, Wolff, and Oates, 1988). These liquid forms contain the bulk of solute and consequently are more concentrated than the parent ice crystal by at least an order of magnitude. In the melt period, wet snow metamorphism occurs. This type of metamorphism is more rapid than dry snow metamorphism. Large grains grow at the expense of small grains in intimate contact with liquid water. Grain clusters dissociate and solute release from grain boundaries is favoured (Davis, 1991). The amount of solute able to diffuse from the grain structure into any discrete meltwater volume depends on the diffusion coefficient for the solute, the time of solute–meltwater contact (determined by flow rate), and the amount of snow leached.

Deeper snow increases the duration of snow–meltwater interaction and gives rise to higher snowmelt concentrations. For example, Marsh and Webb (1979) report

the approximate doubling of initial snowmelt concentrations with the doubling of snow depth. Low melt rates promote the more uniform flow of meltwater through the whole snow matrix and solute scavenging is maximised. Colbeck (1981) showed that slow-moving meltwater picks up solute by molecular diffusion at flow rates $<10^{-7} \text{ m s}^{-1}$. At high melt rates, contact time is less. In addition, preferential flow through vertical macropores also occurs (Kattleman, 1985, 1989), resulting in both a decrease in the snow–meltwater contact time and the mass of snow leached per unit volume of meltwater. Hence, solute scavenging is minimised. The composition of snowmelt thus may vary on a diurnal basis, because solute scavenging is related to the rate of melt (Tsiouris et al., 1985; Tranter et al., 1988b; Bales et al., 1993). Higher concentrations of solute in meltwater are found in the morning and evening, or during periods of shading, when melt rates are lowest.

Generally, meltwater flowing through macropores or flow fingers is more dilute than contemporaneous melt flowing through the snow matrix. The effects of heterogenous flow on fractionation are illustrated in Figure 3.6, where CF is shown for two flow paths, one with the lowest (matrix) and one with the highest (macropore) measured flow. Note that the maximum CF is 10 for the flow path with the lowest flow but only 8.0 for the path with the highest flow. Over the following 6 days, the CF of both flow paths gradually converged until all flow paths had similar values. However, the formation of horizontal macropores in some snow covers due to discontinuities between snow strata, or the impermeability of ice lenses, increases meltwater concentrations because the duration of snow–meltwater contact is increased by horizontal flow (Marsh and Pomeroy, 1993).

The mesoscale distribution of solute in snow cover will also affect the concentration of meltwaters. Discrete snowfalls give rise to snow strata of different compositions. Solute-rich bands also arise from the exclusion of solute from ice lenses formed by the refreezing of meltwater fronts or rain in cold, dry snow. The effect of diurnal melt–freeze cycles is to translocate solute from the surface to the interior or the base of the snowpack. Meltwater in contact with frozen ground refreezes to form basal ice layers, often overlaid with saturated wet snow layers (Marsh, 1990). Thus the result of several diurnal melt–freeze cycles is often to increase the concentration of ions in the first meltwaters issuing from the snowpack (Bales, Davis, and Stanley, 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). However, this may be attenuated by macropore flow that causes meltwater to bypass areas of the solute-rich layers.

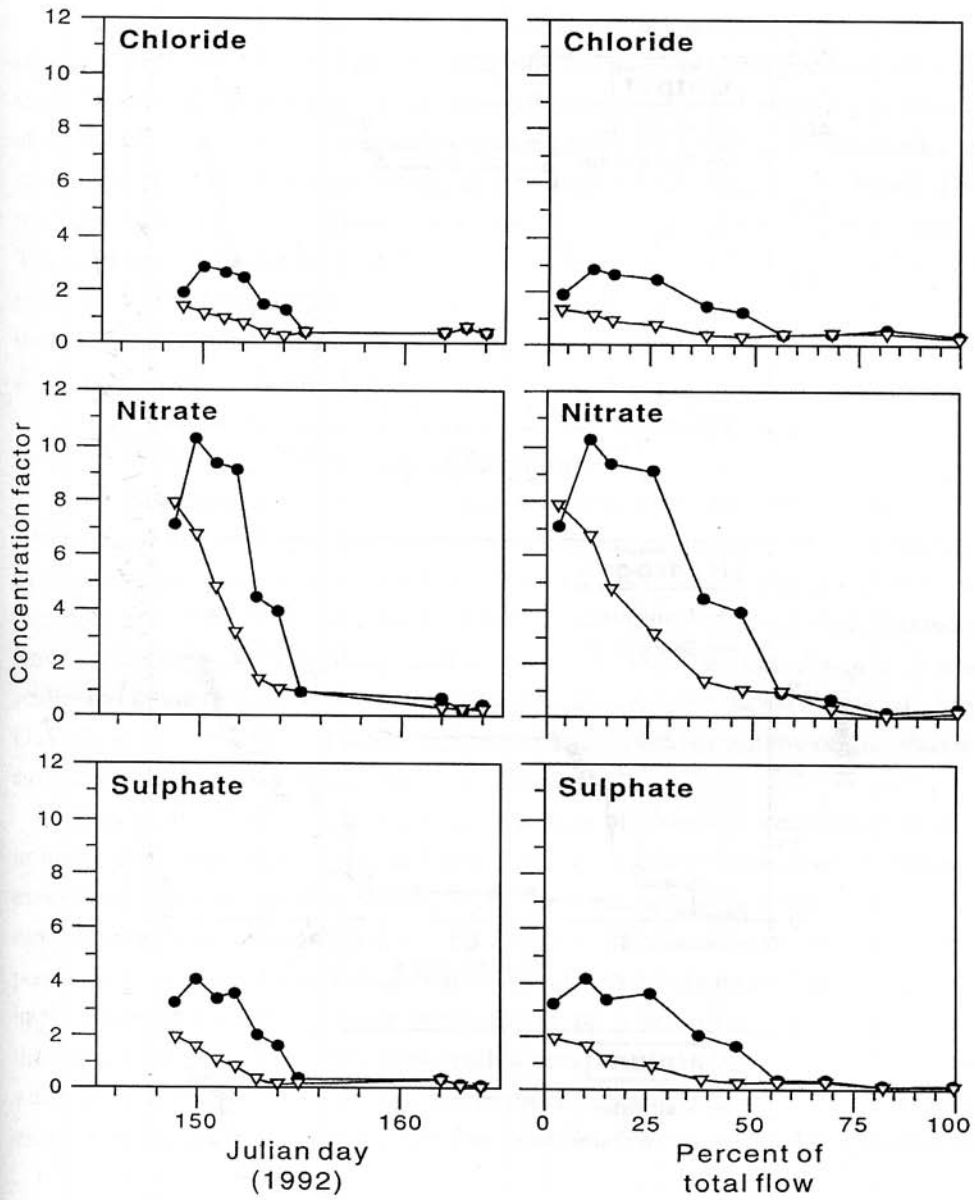
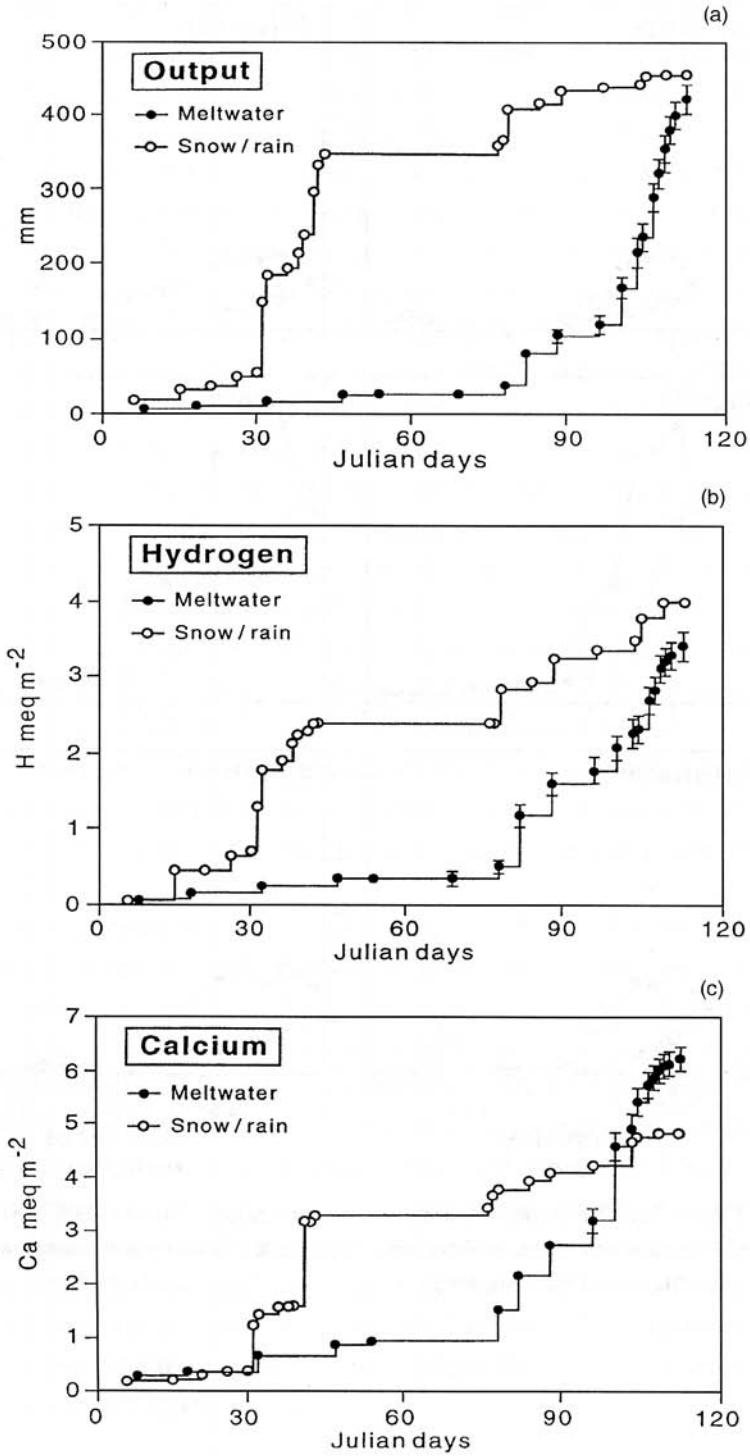


Figure 3.6. The impact of flow rate on the concentration factor of SO_4^{2-} , NO_3^- , and Cl^- in snowmelt. Open symbols denote high rates; closed symbols denote low flow rates (after Marsh and Pomeroy, 1993).



The distribution of solutes in snow and the dynamics of meltwater hydrology increases the difficulty of modeling solute leaching and meltwater composition. However, models for snow meltwater quality have been developed that generate CF as a function of meltwater discharge. These models ignore preferential meltwater flow paths and solute distribution (Goodison, Louie, and Metcalfe, 1986; Stein et al., 1986). They represent bulk models that combine total snow depth and a bulk leaching coefficient, k . The leaching coefficient is based on a first-order removal of the solute from snow by meltwaters (Hibberd, 1984). High values of k give rise to greater values of CF in the initial meltwaters. Values of k vary (e.g., from 0.002 to 0.01 $\mu\text{eq mm}^{-1}$ for NO_3^-), depending on crystal habit and rate of flow.

3.4.2 Snowmelt-Particulate Interactions

Chemical reactions between meltwater and inorganic-organic particles can affect the concentration of solute in meltwater. Chemical weathering of dust scavenged by snow can take place in the atmosphere (Sequeira, 1991; Colin et al., 1987) or within the snow cover (Psenner and Nickus, 1986; Maupetit and Davies, 1991; Puxbaum, Kovar, and Kalina, 1991). Many studies have observed the neutralisation of snow acidity by carbonaceous dusts from a variety of sources, either of local (Colin et al., 1987; DeWalle et al., 1983; Hart et al., 1985; Delmas et al., 1996) or remote origin (Psenner and Nickus, 1986; Loye-Pilot, Martin, and Morelli, 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. Laboratory experiments showed that dust located in the lower strata of snow covers showed more rapid rates of weathering than dust in the upper strata because of increased partial pressures of CO_2 that arise during dust-meltwater interaction (see Figure 3.7). Dust in the lower regions of the snow cover, therefore, was more efficient than dust in the upper strata at neutralizing acidic meltwaters. However, the extent to which this weathering of dusts plays an effective role in the chemical composition of many regional meltwaters has not been truly evaluated or modeled. Delmas et al. (1996) showed that

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Figure 3.7. The neutralisation of meltwater by calcareous dust in the French Alps (after Delmas et al., 1996). (a) The cumulative input of snow water equivalents and rain to the snow cover and the cumulative output of meltwater. 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.

meltwaters from an alpine site in the French Alps with aeolian dust inputs had far lower acidities than those predicted from the acidity of either snowfall or the snow cover before the melt period. Meltwater models that take into account only the bulk ionic composition of snow to compute the acidity of meltwaters thus may overestimate the acidity of meltwaters from snow covers containing mineral dust.

Along with aerosols and dusts, organic debris may be found in considerable quantities in some snow covers (e.g., canopy litter in boreal forest snow covers) (Jones, 1991; Taylor and Jones, 1990). Litter that falls onto snow is relatively unaffected by physical and microbiological activity during the cold accumulation period. However, the leaching of litter by meltwaters removes soluble organics and other chemical species (Fahey, 1979; Jones and Sochanska, 1985; Stottlemeyer, 1987). Surficial ionic exchange also may take place between meltwater and the organic debris (Cronan and Reiner, 1983). Leaching experiments in the field (Barry and Price, 1987; Jones and DeBlois, 1987; Courchesne and Hendershot, 1988) or in the laboratory (Moloney, Stratton, and Klein, 1983; Jones and DeBlois, 1987) with snow lysimeters show, in general, that large amounts of PO_4^{3-} , K^+ , Mn^{2+} , Ca^{2+} , and Mg^{2+} are discharged from litter-laden snow covers and a decrease in the acidity of meltwaters may arise from cation exchange. In contrast, interaction between meltwaters and litter often leads to losses of NO_3^- and NH_4^+ in snow cover because of microbiological activity.

3.4.3 Microbial Activity

During springmelt, the presence of liquid water and the increase in solar radiation stimulates microbiological and invertebrate activity in snow cover. Jones and DeBlois (1987) showed that meltwater production increased microbiological activity on canopy fallout in forest snow cover. The presence of meltwater also results in photosynthetic activity of truly motile algal populations within the snow cover (Hoham, 1987; Hoham and Duval, Chapter 4). 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 $0.67 \text{ eq[N] ha}^{-1} \text{ day}^{-1}$ and $1.05 \text{ eq[N] ha}^{-1} \text{ day}^{-1}$, respectively (Gamache, 1992; Hoham et al., 1989). The decreases sometimes may be masked by dry deposition of the same species from the atmosphere. Jones and Sochanska (1985) showed that the losses (photosynthesis) or gains (dry deposition) of NH_4^+ or NO_3^- in the snowpack were related to the water content of the snow cover. High amounts of free water in snow cover lead to losses of NH_4^+ and NO_3^- , whereas NH_4^+ and NO_3^- concentrations increased when low amounts of free water were present. Low amounts of free water lower algal activity and the rate

of dry deposition of the N species to snow may then become higher than the rate of assimilation by algae. The loss of nutrients in snow meltwaters over the whole melt season may be appreciable, approaching 20–30 percent in some years (Jones, 1991). However, there is considerable annual variation due to annual variations in snowmelt climatology (Jones and DeBlois, 1987).

Laboratory studies on melting snow containing litter fallout from forest canopy (Jones and DeBlois, 1987; Jones and Tranter, 1989) and cultivated snow algae (Gamache, 1992) confirm the losses of nutrients observed in the field. NH_4^+ and NO_3^- depletion rates in meltwater simulators varied between $0.05 \text{ eq[N] ha}^{-1} \text{ day}^{-1}$ (Gamache, 1992) and $0.42 \text{ eq[N] ha}^{-1} \text{ day}^{-1}$ (Jones and Tranter, 1989) for NH_4^+ and $0.86 \text{ eq[N] ha}^{-1} \text{ day}^{-1}$ (Gamache, 1992) and $1.25 \text{ eq[N] ha}^{-1} \text{ day}^{-1}$ (Jones and Tranter, 1989) for NO_3^- . The total nutrient loss ($\text{NH}_4^+ + \text{NO}_3^-$) may represent up to 36 percent of the nitrogen available in the snow before melt (Gamache, 1992). The addition of coniferous forest litter to snow containing cultivated algae leads to increased depletion rates of $0.62 \text{ eq[N] ha}^{-1} \text{ day}^{-1}$ for NH_4^+ and $2.22 \text{ eq[N] ha}^{-1} \text{ day}^{-1}$ for NO_3^- in similar laboratory experiments. The total N loss, $2.8 \text{ eq[N] ha}^{-1} \text{ day}^{-1}$, was equivalent to 62 percent of the total N available in the snow before melt (Gamache, 1992).

These measured depletions of total N in both laboratory and field studies (see Figure 3.8) are consistent with reported rates of algal biomass production in snow covers. Mosser, Mosser, and Bruck (1977) measured fixation of ^{14}C in Montana snow fields

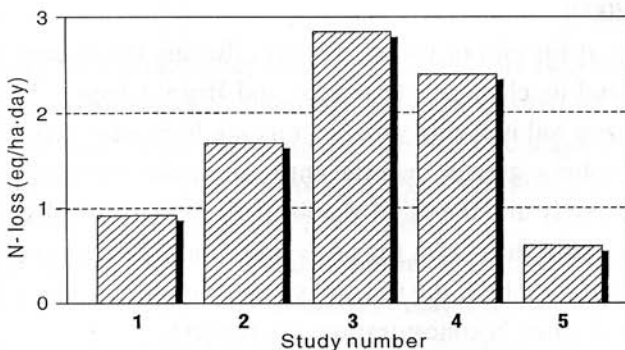


Figure 3.8. Nutrient (N) losses for field and laboratory experiments on snow/meltwater systems. Bars: 1, laboratory study (snow with added algae; Gamache, 1992); 2, laboratory study (snow with added coniferous litter; Jones and Tranter, 1989); 3, laboratory study (snow with added coniferous litter and algae; Gamache, 1992); 4, field study (Mosser, Mosser, and Brock 1977); 5, field study (Thomas, 1994).

and calculated an approximate depletion rate of $2.4 \text{ eq[N] ha}^{-1} \text{ day}^{-1}$ based on C:N ratios for algal biomass. A similar study by Thomas (1994) on fixation of ^{14}C by snow algae in the Sierra Nevada gave values that are equivalent to depletion rates of $0.6 \text{ eq[N] ha}^{-1} \text{ day}^{-1}$. Thus rates of N losses measured directly in the field, simulated in the laboratory, and calculated from C-fixation rates in situ are consistent. They show that algal productivity can deplete up to two-thirds of the total inorganic N load in snow covers before the snow cover is completely melted.

In contrast to these snow covers, which have a significant input of organic matter, microbial activity may be significantly reduced in snow cover far from inputs of degradable litter. Jones, Duchesneau, and Handfield (1994) determined that the numbers of microorganisms within the snow cover of a High Arctic ice cap that underwent intermittent melting during the summer were not sufficient to significantly affect the measured concentrations of inorganic N in the snow.

3.4.4 Basal Processes: Soil–Meltwater Interactions

The release of liquid water and soluble nutrients from melting snow cover leads to significant changes in the rate and type of microbiological activity in the soil (Brooks, Williams, and Schmidt, 1997) and greater fluxes of gases to the snow cover (Brooks, Williams, and Schmidt, 1996). However, the subsequent diffusion of the gas through the snow may be counteracted by the dissolution of gas in the percolating meltwaters and consequent transport back into the soil water and groundwater systems (van Bochove et al., 2000).

Soil may be frozen at the end of winter, greatly affecting the volume and timing of meltwater runoff and its chemistry (Pomeroy and Brun, Chapter 2). Infiltration of meltwater into frozen soil is highly variable, ranging from near zero (i.e., the soil is impermeable) to a volume greater than the entire snowpack water equivalent (i.e., all meltwater enters directly into the soil) (Marsh, 1990, 1991). Infiltrating meltwater refreezes if the soil is sufficiently cold, thus delaying runoff of some snowmelt. As a result, the availability of N to both the soil and stream runoff may be limited during the initial runoff period when N concentrations are highest.

3.5 Snow Cover Nutrient Fluxes and Ecosystem Budgets

For most terrestrial ecosystems, nutrient input from the atmosphere is the major source for total pools of certain nutrients (e.g., N, S), while export by gaseous

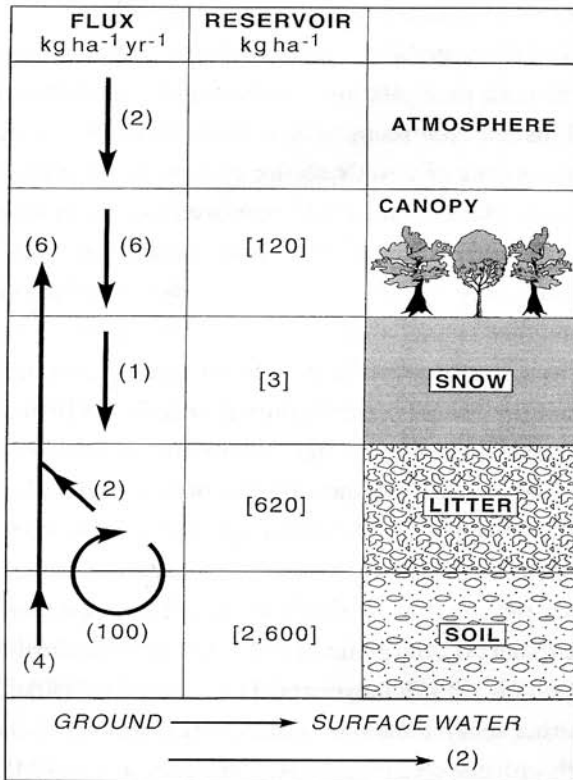


Figure 3.9. Simplified N pools and fluxes in a boreal forest (after Jones, 1991).

emissions back to the atmosphere and aquatic transport away from the system constitute the major outputs in ecosystems that are snow covered for much of the year. The major input arises in snow meltwaters in spring. The major output also may occur during the same period, when hydrological export by streams is the greatest because of the greater meltwater runoff (Williams, 1993).

On an annual basis, the input and/or output of certain key nutrients are relatively small with regard to the amount of nutrient that is being recycled within the system itself. This is particularly true in the case of nitrogen (see Figure 3.9), which is a limiting factor for growth in many terrestrial ecosystems. For example, an atmospheric input to a typical boreal forest of 0.15 keq N (2 kg) ha⁻¹ year⁻¹ (1.5 kg ha⁻¹, wet deposition; 0.5 kg ha⁻¹, dry deposition) represents only 0.2 percent of the N pool in the above-ground vegetation and forest floor (733 kg ha⁻¹; 116 kg ha⁻¹, above ground vegetation; 617 kg ha⁻¹, forest floor) and only 0.001 percent of the total pool of 3,350 kg ha⁻¹, including mineral soil horizons. As N fixation is relatively low in

boreal forests, N cycling involves mostly intrasystem transformations. However, as only about 10–20 percent of the vegetative and forest floor N pool is involved in the annual turnover of the nutrient, precipitation inputs can contribute up to 1 percent of the annual turnover. This may not seem to be a major factor, but precipitation is one of the few long-term sources of new N to the system. In addition, the impact of the precipitation input on plant productivity may well outweigh its quantitative contribution to the N pool. This is because the ionic N species (NO_3^- , NH_4^+) in precipitation are the only major forms of N actively absorbed by plants (Haynes and Goh, 1978) and thus are readily bioavailable.

In the case of boreal forests, snow covers the ground for up to 6–7 months of the year and can represent up to half the annual precipitation (Pomeroy and Brun, Chapter 2). During the spring melt period, meltwaters deliver nutrients to the forest floor and, in spite of some nutrient losses of N from snow during both winter and spring, meltwaters can discharge up to 2–4 kg ha⁻¹ of N within a period of a few weeks. However, there is little knowledge of the impact of such a relatively massive discharge of readily bioavailable N over a short time period on the N cycle of the forest ecosystem. It is known, however, that the export of inorganic N (NO_3^-) is greatest during the runoff period (Jones and Roberge, 1992; Stottlemeyer and Toczydlowski, 1990). This has also been found to be true for other ecosystems such as hardwood forests (Rascher, Driscoll, and Peters, 1987) and high-altitude alpine sites (Williams, Brown, and Melack, 1993). Typical values for the export of N (NO_3^-) by streamwaters during melt in a boreal forest are 0.65 kg N ha⁻¹ (Skartveit and Gjessing, 1979) and 1.7 kg N ha⁻¹ (Jones and Bedard, 1987). In general, the export of N as NH_4^+ is very low as stream concentrations of NH_4^+ are negligible. However, a significant part (55 percent) of total N export in the form of NH_4^+ has been reported before snowmelt during low-flow regimes in a stream draining a snow-covered mountain watershed (Lewis and Grant, 1980). In all studies, the export of total N is small compared with the amount of N cycling in the system.

The export of N as NO_3^- originates both from the solute in the meltwaters (Williams, 1993) and/or from the leaching of the species from soil after overwintering 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 in winters when the soil was frozen. They suggested that soil frost may inhibit denitrification or any other biological sink for NO_3^- and/or increase nitrification. The hypothesis about blocking denitrification seems to be supported by the work of Christensen and Tiedje (1990), who found that thawing of soils during spring resulted in very large increases in emissions of N_2O to the

atmosphere. However, the suggestion that soil frost increases nitrification is not supported by the study of Stottlemeyer and Toczydlowski (1990) on an upper-Michigan watershed. In this field experiment, the data from soil lysimeters under snow cover indicated that nitrification occurs throughout the winter in the upper organic soil horizons because of above-freezing temperatures. Slow sustained meltwater discharge moved the mineralized nitrogen to lower inorganic horizons where it was not taken up because of low biological activity. Some of the NO_3^- was then removed during the main melt period by macropore flow into surface water channels.

The studies described in this section have addressed only certain aspects of N cycling under snow. In a recent study on the influence of snow on the nutrient dynamics of alpine ecosystems, Brooks et al. (1996) attempted to quantify N budgets by determining the input of N due to snowmelt, the export of N by stream waters and gaseous emission (N_2O), and overwinter N mineralisation in the thawed soil beneath the snowpack. They reported that the inorganic N input (NO_3^- , NH_4^+) from snow represented only 3–6 percent of the net N mineralisation, showing that intrasystem cycling was the main source of inorganic N available for the start of the growth season. They also estimated that the winter/spring N losses by denitrification (N_2 , N_2O) were equal to the NO_3^- -N input by snowmelt and represented 50 percent of the annual gaseous N loss. In this particular system, the loss of N via runoff was negligible during snowmelt. This latter result contrasts the hydrological losses measured by Williams (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 different responses of ecosystems to the input of N by snowmelt. The factors controlling the hydrological export of N as NO_3^- are poorly known and remain to be elucidated by future research.

3.6 Summary and Future Research Needs

Recent studies of the chemistry of snow covers show that snow is not a passive reservoir of the chemical species scavenged from the atmosphere during snowfall. Snow-atmosphere exchange by dry deposition and volatilisation, accompanied by various physical processes such as air 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 also take place and the presence of microorganisms influences nutrient concentrations during the melt period. Snow covers often also show strong concentration gradients of CO_2 and trace gases as snow mediates the gaseous emissions from the soil to the atmosphere.

Some of these processes are understood well enough for models of the chemical evolution of snow cover to be constructed. However, for most of the processes described we have very little knowledge or data on which to base any viable models. This is particularly true for the evolution of chemical species during wind-blown snow events and dry cold snow metamorphism. There is practically no knowledge at all of photochemical reactions – even though they are thought to play a significant role in the chemistry of some snow covers. Finally, the factors that control the microbiological activity in snow and the effect of vegetative cover on snow cover composition are only now being brought to light. We need, above all, well-designed laboratory and field experiments that will permit us to acquire the data and knowledge to gradually construct the relationships that exist between snow and snow-covered ecosystems.

Thus, the relationships between the chemistry of snow, terrestrial nutrient cycles, and ecology will continue to be elucidated. Much remains to be done. We hope that we have given the reader an insight into the processes that form the basis of the chemical dynamics of snow, and more importantly, we hope that we have stimulated interest about the fascinating study of life in snow and snow-covered ecosystems. These subjects are further developed in the following chapters.

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