Wind-blown Snow: Sublimation, Transport and Changes to Polar Snow

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Keywords

Abstract

Accumulation of snow and ions in polar regions is a residual of the blowing snow phenomenon, including sublimation. Ion accumulation depends on the blowing snow regime and occurs in two modes of retention and resuspension. Ion deposition in polar snow is therefore inherently unstable. An understanding of wind speed regimes is required to predict ion concentration and annual retention in wind-blown snow. This adds considerable complexity to attempts to interpret the chemistry of recent ice cores and to calculate glacial mass balances.

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INTRODUCTION

In their hallmark work on the Antarctic blowing snow phenomenon, Budd et al. (1966) recounted a quote of Peary (1898) that still has relevance to interpretation of glacial phenomena:

> "There is one thing of special interest to the Glacialist - the
> transport of snow on the ice-cap by the wind....."

Following Peary's reasoning, the blowing snow phenomenon was studied intensively in the Antarctic several decades ago (Lister, 1960; Mellor and Radok, 1960; Dingle and Radok, 1961; Budd et al., 1966) with more recent studies by Kobayashi (1978), Takahashi (1985) and Moore et al. (1994). Other work of similar nature has been conducted in the Arctic (Dyunin, 1959; Benson, 1982; Tabler et al., 1990b; Benson and Sturm, 1993). Blowing snow is quite frequent in the Antarctic, Dalrymple (1966) noted snow transport occurred from 30-55% of the time in the South Pole region, increasing to 55-65% of the time at Byrd Station. Fujii (1981) found snow accumulation to occur in only one or two out of every three years in East Antarctica because of wind erosion and sublimation. At Mizuho Station, East Antarctica, Takahashi et al. (1994) estimated that of an annual snowfall of 140-260 mm water equivalent, 100 mm/year is eroded from surface snow and transported away by blowing snow and 50 mm/year sublimates. The invariable conclusion is that snow covers in the polar regions are dramatically transformed by wind redistribution; it may therefore follow that the chemistry of

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these snow covers is, as well, altered by wind redistribution. In affirmation, Delmas and Jones (1987) and Pomeroy et al. (1991a, b) measured and modelled changes to snow chemistry during drifting in temperate locations, Pomeroy et al. (1993b; 1995) presented evidence of chemical alteration of wind-blown snow in the Canadian Arctic and Osada (1994) showed strong chemical fluctuations in wind-blown snow for the Antarctic. Redistribution leads to a re-evaluation of our concepts of snowcover formation; in polar regions snow does not normally fall in a simple fashion from the atmosphere to the surface to be buried by subsequent snowfalls, but more often falls and moves horizontally, carried by the wind, to be either sublimated, resuspended or deposited. Deposition onto the snowpack occurs when and where the atmospheric surface shear stress is insufficient to continue transport. After initial deposition the snow may be eroded and transported again. In extreme blowing snow events some snow may be lifted by turbulence tens or hundreds of metres into the atmosphere where sublimation reduces its mass to insignificance. During transport the parent snow forms a turbulent mixture with the atmosphere, aerosols and snow from other sources, hence it may be mixed with older or fresher snow, is exposed to the atmosphere's chemical constituents and undergoes relatively rapid sublimation. These processes add considerable complexity to attempts to relate the chemistry of the resulting surface snow to the parent snowfall as outlined by Neftel (1996) and Bales and Choi (1996) in this volume.

Effects of wind redistribution that are important to ice core chemistry may be classified as concentration effects and deposition effects, as they affect the chemical concentration of, and thickness of, annual firn layers.

Concentration changes in snow occur during:

    a) sublimation of ice where chemical species are conservative,

    b) scavenging (adsorption) of particulates or vapours by the surface of blowing snow particles,

    c) volatilization of chemical species unrelated to sublimation effects.

Deposition of snow may be concomitant with:

    a) volatilization of chemical species,

    b) snow deposition fluxes that are not equal to the snowfall flux,

    c) co-deposition of atmospheric constituents.

Mass, ion and energy balances for small control volumes (i.e. over fetches of 1 m to 1 km) are used to calculate changes in concentration or deposition (Pomeroy et al., 1991a). However

the application of control volumes to large areas such as the Antarctic ice sheet has not been explicitly examined, though it has been proposed for the Arctic by Pomeroy et al. (1995).

This paper reviews recent developments in our understanding of chemical changes associated with wind-blown snow. It also reassess the current conceptualisations of precipitation and accumulation in polar regions, discusses the implications for the chemistry of ice cores and attempts to link the chemistry of these ice cores with that of the atmosphere.

PHYSICS

The physical processes during wind transport and snowcover formation are extremely important for understanding the concomitant chemical transformations. This section will briefly review the physics associated with formation of snowcovers in windy, exposed locations. For recent comprehensive reviews of the phenomenon the reader is referred to Tabler et al. (1990a), Pomeroy et al. (1993a) or Pomeroy and Gray (1995).

Snowfall and Meteorological Measurements

Any discussion of the physics of snow accumulation in polar regions would be incomplete without addressing measurement problems. In extreme polar environments there are few locations where, year-round, sufficient and appropriate measurements are taken to quantify true snowfall and where wind speed, temperature, humidity and the time of occurrence of blowing snow are available for use in blowing snow transport, sublimation and accumulation calculations. The situation is particularly discouraging in central Greenland and the interior of Antarctica where even the direction of the current mass balance is not known (Houghton et al., 1990). Recent models of precipitation predict *snow accumulation* not *snowfall* for polar regions (Connolley and Cattle, 1994). Verification of climate models is conducted against snow accumulation, which is normally measured as an increase in snow depth on the ground instead of true snowfall (Marshall et al., 1994). In windy environments the difference between snowfall and snow accumulation is substantial and may vary more or less by an order of magnitude between exposed and sheltered locations. Where precipitation gauges are used, an

undercatch due to wind exposure results (Goodison, 1981). This undercatch increases with wind speed; where mean wind speeds are normally in excess of 5 m/s as in the polar regions, an undercatch of over 50% is expected for unshielded gauges. Further undermeasurement, where snow collection cylinders are emptied routinely, is due to non-recording of trace snowfalls and to wetting loss of snow that is retained in the cylinder. Benson and Sturm (1993) suggest that Arctic Alaskan snowfall measurements are underestimated by a factor of three. When Goodison's techniques were used to correct winter precipitation records for Resolute, Northwest Territories, Canada, annual snowfall increased by 64% to 161% (Pomeroy and Goodison, 1995). Kobayashi et al. (1985) estimated snowfall during blowing snow by examining asymptotes of the vertical profile of blowing snow in East Antarctica at Mizuho Station. Their estimated snowfall fluxes during blowing snow ranged as high as 0.15 mm/hour. In regions of the Antarctic subject to katabatic snow-storms this flux results in annual snowfall rates during blowing snow ranging from 9.6 to 720 with an average of 140 mm/year. Because of the high wind speeds this snowfall goes largely unrecorded by standard precipitation gauges.

To calculate hourly blowing snow fluxes, wind speed, air temperature, humidity, corrected snowfall, depth of snow on the ground and the time of occurrence of blowing snow are needed (Pomeroy et al., 1993a). Monthly fluxes may be estimated using a simpler model that does not require the time of occurrence of blowing snow (Pomeroy and Gray, 1994). Unfortunately unattended anemometers in polar regions are subject to rime and snow-filled cups causing them to underestimate wind speed. Such data can result in serious errors in blowing snow calculations. The most usable data for blowing snow calculations is that collected at manned polar stations where hourly visual observations and frequent instrument inspections/corrections are made.

Wind Transport of Polar Snow

As snow is transported by wind, the crystals undergo changes to their shape and size and upon re-deposition form drifts and surfaces of higher density than the original snow. Crystals are abraded by particle impact, shattered and sublimated, to give oblong amorphous shapes with characteristic size distributions even in cold, polar environments (Budd, 1966; Schmidt, 1982).

Wind transport of snow is closely linked to synoptic winds in the Antarctic. Kodama et al. (1985) showed that the katabatic wind dropping off the ice-sheet is partially sustained by the additional weight of blowing snow in the air-flow and the cooling of the air by sublimation of blowing snow. A positive feedback develops where as more snow is entrained, the katabatic wind speed increases and the erosive force increases. Wind transports snow in two substantive modes: saltation and suspension.

Saltation - particles which move by a skipping action along the snow surface form this two-phase flow. Particle trajectories follow a near-vertical ascent with a flat descent; initial horizontal velocity is quite low but the final horizontal velocity exceeds the ambient wind speed. Saltating particles attain a maximum trajectory height of from 0.1 to 5 cm, increasing with the square of the friction velocity. The snow cover comprises a "bed" of snow from which particles are entrained and deposited and over which saltation occurs. This bed is a mixture of bonded crystals metamorphosed by particle impact and vapour transfer. Particles are normally ejected from this bed as part of a "splash" from the impact of a descending particle. In order to be ejected by splash the energy of impact must be sufficient to overcome cohesion and the work required to break bonds of surrounding and underlying particles. Over a uniform surface, erosion of snow particles occurs when the shear stress exerted by the wind, τ, exceeds the surface shear strength, τ_s where,

$$\tau_p = \tau - \tau_s \tag{1}$$

and τ_p is the shear stress available to drive saltation. Evidence of the effect of varying shear stress on snow erosion in the Antarctic is presented by Takahashi et al. (1994); along a 850 km transect of snow surveys, snow erosion reduced the accumulation rate over large-scale convex surfaces where shear stress is at a maximum. The atmospheric shear stress, τ is equal to $\rho_a u^{*2}$, where ρ_a (kg/m^3) is atmospheric density and u* (m/s) is the friction velocity and found over blowing snow as,

$$u* = \frac{u_z \, k}{\ln(\frac{C \, z}{u*^2})} \tag{2}$$

k is von Kármán's constant (0.4), u_z is the wind speed (m/s) at height z (m) and C is an experimentally-derived constant found to be 163.3 m/s^2 (Pomeroy and Gray, 1990). As a

result of this energy balance and measurement of saltation efficiency, Pomeroy and Gray related steady-state saltation transport to the wind speed in excess of the threshold level at the beginning or cessation of transport. Typical threshold wind speeds (10-m) range from 6 to 9 m/s for wind-hardened snow. The transport rate of saltating snow increases approximately linearly with the wind speed and at high wind speeds comprises less than 10% of the total transport rate.

Suspension - when the upwards drag on a saltating snow particle becomes greater than the gravitational force the particle becomes suspended and moves along turbulent flow lines at a mean horizontal velocity close to that of the air. Suspended snow derives from saltating snow or falling snow, never directly from the surface. Budd et al. (1966) extensively measured suspended mass flux of snow in Antarctica, describing the particle size distribution and the decrease in particle size with height. The layer of suspended snow reaches from the top of the saltation layer to up to several hundred's of metres, increasing with upwind fetch of blowing snow. The concentration of suspended snow is extremely sensitive to wind speed and declines exponentially with height, from nearly 800 g/m^3 near the top of the saltation layer to less than 0. 01 g/m^3 at several metres height. Pomeroy and Male (1992) developed a turbulent diffusion model of suspended snow, whose results agree with Budd et al. The model demonstrates that the suspended transport rate dominates the mass transport of blowing snow and increases with roughly the fourth power of wind speed.

The total transport rate of snow is a compilation of transport in its three modes and is very sensitive to the upwind fetch distance and to snow supply. A recent practical formulation for the downwind transport rate of snow, Q_T as a function of wind speed in polar regions is that by Tabler et al. (1990a). Tabler et al.'s formulation is derived from profiles of Antarctic mass flux measurements published by Mellor and Fellers (1986), integrated to a height of 10 m, and coupled with the mass concentration limitation suggested by Pomeroy and Gray (1990) for the saltation layer. The integral height of 10 m and lack of a characteristic threshold transport value are not important limitations for polar regions where upwind fetches are long and persistent snowcover may be presumed. Tabler et al.s expression has the form,

$$Q_T = 0.0000014 \ u_{10}^{4.2} \ . \tag{3}$$

As shown in Fig. 1, this transport rate increases dramatically with wind speed; given a 10-m wind speed of 8 m/s the transport rate is 0.0087 kg/s per metre width, but with a wind speed of 20 m/s the transport rate is 0.408 kg/s per metre width. Because of the non-linear increase in transport with wind speed this equation should not be used with long-term average wind speeds to estimate mean transport rates; much of the annual transport may be due to a few extreme events.
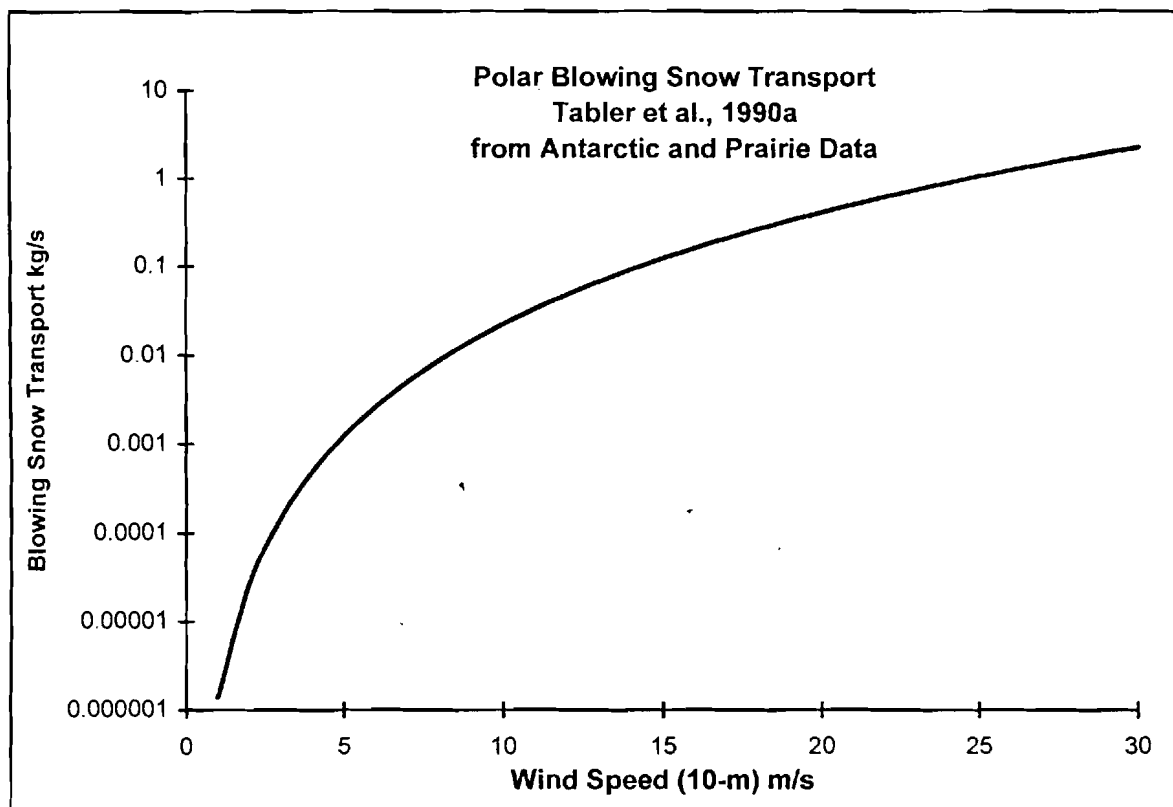
Figure 1. Blowing snow transport rate as a function of wind speed for continuous, level snow covers (after Tabler et al., 1990).

Sublimation of Polar Snow

Stationary Snow Sublimation. Sublimation of snow is driven by inputs of atmospheric sensible, Q_H, and net radiative, Q^*, heat, which must be sufficient to drive the latent heat requirements of the sublimation process. For cold, level and complete snowpacks over deep snow fields the energy balance for snow may be written as,

$$Q_E = Q^* + Q_H + Q_S \qquad\qquad (4)$$

where Q_E is the latent heat flux. Q_S is the vertical heat flux at the snow surface and takes into account internal energy changes and any sensible heat flux from the deep polar snow. For polar snows Q_S is normally quite small and because of high albedo Q^* is usually negative or slightly positive except in the warmest polar conditions. The fluxes of latent and sensible heat to the surface are controlled, in analogy to Fick's Law, by the gradients of wind speed, air temperature and humidity above the surface and the relevant turbulent diffusivities of momentum, heat and water vapour (Male, 1980). The turbulent diffusivities for heat and water vapour are normally related to the diffusivity for momentum. Granger and Male (1978) showed that for slightly-stable conditions over snow the turbulent diffusivity for heat is approximately equal to that for momentum and decreases to eventually equal half that for momentum as stability increases. In contrast they showed that the diffusivity of water vapour is roughly constant at half that of momentum for all stable conditions. Turbulent diffusion of water vapour from or to a stationary snow surface is limited by non-existent turbulence at the snow surface and the relatively small surface area of ice exposed to the wind. Wind-pumping may increase these values above previous estimates but sublimation rates are limited by the available energy (Albert and McGilvary, 1992). For polar snows, the small air temperature gradients and low rate of turbulent transfer result in the latent heat flux tending to correspond to the net radiation flux. This is analogous to cold, continuous prairie snowcovers where Male and Granger (1979) reported daily sublimation ranging from 0.02 to 0.3 mm SWE/day with a mean of 0.1 mm-SWE/day in pre-melt conditions. Most sublimation during high sun periods was counter-balanced by frost accumulation in the evening. The values reported by Male and Granger are for a higher energy environment than is normally the case for polar snow and should be considered an upper limit to sublimation from "in-situ" snowcovers. The constant exposure of polar snow can result in appreciable cumulative in-situ

sublimation however. Fujii and Kusunoki (1982) measured sublimation of 50 mm/year at Mizuho Station using an ice-filled evaporation pan.

Blowing Snow Sublimation. In contrast to stationary snow, blowing snow has a high surface area to mass ratio and is subject to intense atmospheric turbulence, hence the potential for turbulent transfer of heat and water vapour is quite high. Pomeroy (1989) shows that sublimation from a column of blowing snow at air temperatures just below the freezing point, low relative humidity (70%) and high wind speed (25 m/s or severe blizzard condition) can reach 18 mm SWE/hour, well in excess of the maximum in-situ sublimation rates. Because of the small size and high albedo of blowing snow particles, their net radiative flux is small and can usually be neglected in sublimation calculations. For a single particle the energy balance is therefore one between latent and sensible heat, controlled by sensible heat transfer to the particle surface and a thermodynamic equilibrium. Pomeroy et al. (1993a) operationalised the sublimation calculation by assuming that particles remain at the ice bulb" temperature and instituting appropriate vertical profiles of particle size distribution, mass concentration (from saltation and suspension calculations), turbulent velocity, air temperature and humidity in the calculation procedure. The resulting sublimation rates for a column of blowing snow are shown in Fig. 2 for a variety of temperatures, humidities and wind speeds. The example shows overestimates sublimation in certain polar environments where temperatures are regularly lower than -30° C and relative humidities exceed 90%.

Accumulation of Polar Snow

Many conceptualizations presume that the accumulation flux is equivalent to the snowfall flux in polar regions (Connolley and Cattle, 1994; Marshal et al., 1994) however it is in fact a residual of snowfall, sublimation and blowing snow processes. The instantaneous snow accumulation flux, $Q_{surface}$, is calculated using a mass balance of snow fluxes to a control volume over the surface of interest (Pomeroy et al., 1991a) where

$$Q_{surface} = \frac{dQ_T}{dx}(x) - Q_E + Q_{snowfall} \qquad (5)$$

The change in transport fluxes over horizontal distance x is an important term when fetches are short, terrain uneven or flow disturbed by buildings, etc. and can be positive or negative depending upon increasing or decreasing transport with distance. The transport change term becomes insignificant over the large homogeneous fetches typical of polar regions. Q_E is the sublimation flux expressed in $kg/(m^2s)$ as are all other fluxes and $Q_{snowfall}$ is the instantaneous snowfall flux. Note that $Q_{surface}$ can be positive or negative, even when the snowfall flux is large. The quantities frequently measured in polar regions are $Q_{surface}$ and more rarely $Q_{snowfall}$ (corrected for gauge loss). A two-phase flow model such as proposed by Pomeroy et al. (1993a) is required to fully evaluate Q_E for short time periods.
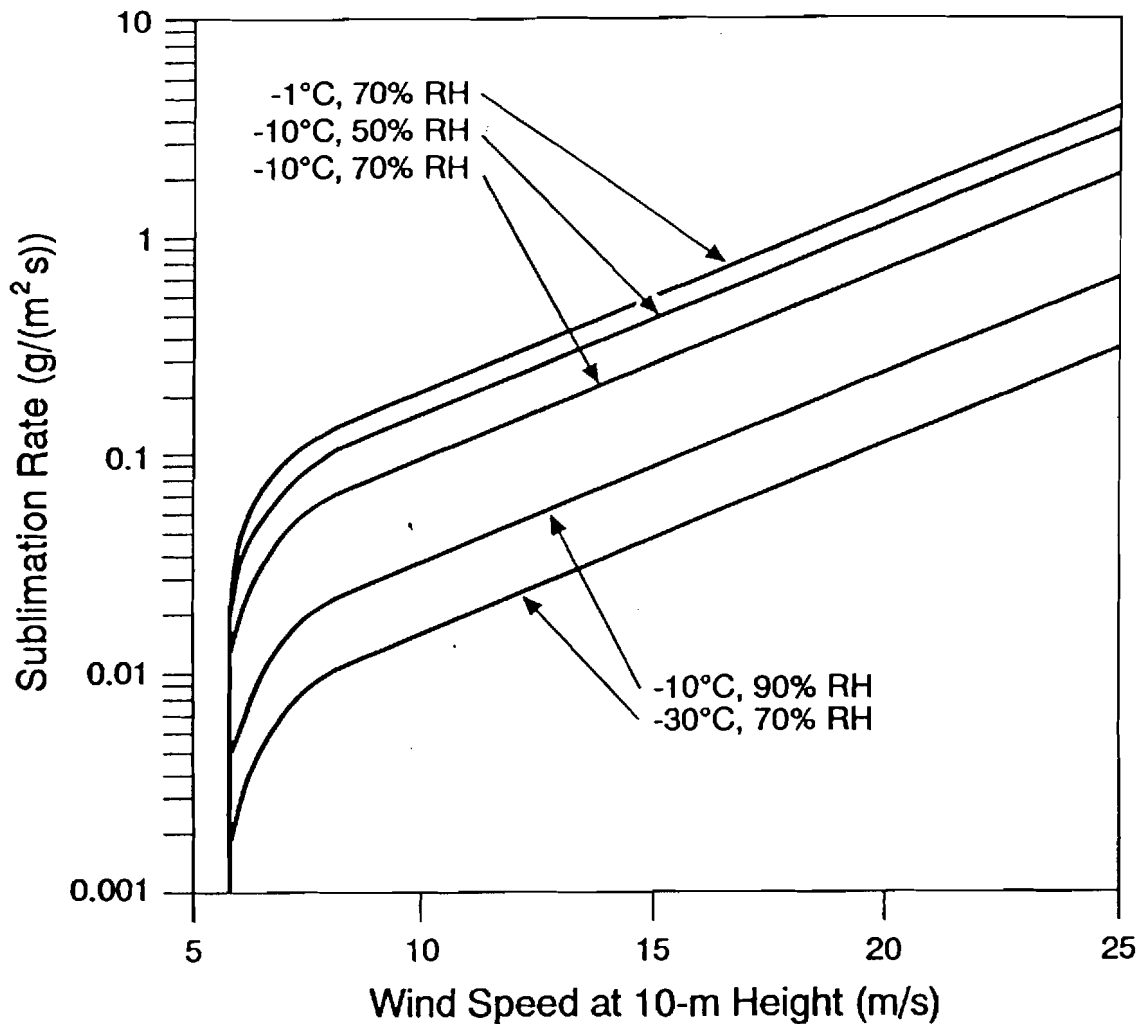

Figure 2. Blowing snow sublimation rate as a function of wind speed, air temperature and humidity for a column of blowing snow over a continuous, level snow cover (after Pomeroy et al., 1993).

Where detailed, suitable meteorological information is not available, a monthly climatological model can be used to estimate the monthly transport, sublimation and hence accumulation (Pomeroy and Gray, 1994). The model presented here was developed using data from smooth snowcovers from more temperate conditions (monthly mean of daily minimum air temperature -26.5 to -0.6 $^\circ$C) than found in polar regions, though it has been verified for seasonal snowcovers in the Canadian Arctic (Pomeroy et al., 1995). For polar regions one can assume that melt is negligible and that in-situ sublimation fluxes are usually small. In this application a uniform snow covered fetch of 1 km is assumed, though this fetch length will be shorter than many fetches on the major ice sheets. Following Pomeroy and Gray (1994), the monthly transport of snow, Q_T(monthly) (mm SWE) from a 1-km fetch, ($-dQ_T/dx$ (x)) is found as,

$$Q_T(monthly) = -14.33 + 2.257\ u_{10} - 0.245\ T_{max}$$
$$+ 0.046\ RH_{max} + 0.0786\ P_m$$

(6)

where u_{10} is the means monthly wind speed (m/s), T_{max} is the monthly mean of daily maximum air temperature ($^\circ$C), RH_{max} is the monthly mean of daily maximum relative humidity (%) and P_m is monthly snowfall (mm SWE). The monthly sublimation of snow, Q_E(monthly) (mm SWE) from a 1-km fetch, is similarly found as,

$$Q_E(monthly) = 7.206 + 1.764\ u_{10} - 0.158\ T_{max}$$
$$- 0.176\ RH_{max} + 0.191\ P_m$$

(7)

Equations 6 and 7 were developed to express monthly fluxes in mm SWE for a 1-km fetch; the sum of the two monthly fluxes does not vary substantially for fetches greater than 0.5 km. For fetches other than 1 km Q_T is divided by the fetch distance (km), and any difference between that and the 1-km value is added to the sublimation flux. Regardless of fetch length, the combination of these equations can provide the monthly accumulation flux in mm for polar environments where,

$$Q_{surface}(monthly) = 7.124 - 4.021\ u_{10} + 0.403\ T_{max}$$
$$+ 0.13\ RH_{max} + 0.73\ P_m$$

(8)

The third term on the right hand side of Eq. 8 indexes the frequency of wet snow for mean temperatures from -28° to $0^{\circ}C$ and has no physical meaning at temperatures below this range; therefore a constraint is applied to T_{max} in polar regions where $T_{max} \geq -28^{\circ}C$. This model is not sensitive to fetch distance when that distance is greater than 300 m and presumes that monthly snowmelt and in-situ sublimation are negligible. For the extremely long fetches of the central Antarctic and Greenland ice sheets any difference between the accumulation predicted by Eq. 8 and precipitation may be presumed to have sublimated in transit. Figure 3 shows the monthly snow accumulation as a function of wind speed for a range of air temperatures and relative humidities and a monthly snowfall of 30 mm SWE. For typical polar "winter" conditions (maximum daily maximum temperatures less than $-25^{\circ}C$) less than half the monthly snowfall of 30 mm accumulates when the mean monthly wind speed exceeds 4 m/s and net snow erosion occurs when mean wind speed exceeds 8 m/s. This has important implications for relating the annual snow accumulation in ice cores to the precipitation record. For instance snow might be deposited, erode months later and be redeposited in a mixture with snows of differing ages. If the chemical signature of such snows vary, then the chemistry of the final deposited layer will be more difficult to relate to specific atmospheric conditions.

CHEMISTRY

Changes in the chemical composition of wind-blown snow result from unequal fluxes, including transformations, of solute and solvent, with respect to the snow particle. Changes in the annual deposition, or load, of chemicals in snow result when the flux of chemicals, including transformations, to or from a surface snowcover is caused by blowing snow. Several potential changes to wind-blown snow chemical composition will be examined, they relate to two important aspects of the blowing snow phenomenon.

i) *Blowing snow is well-exposed to the atmosphere.* A 1-m thick layer of surface snow of average polar density (300 kg/m^3) has an internal surface area of 21.7 m^2/kg (Perla, 1985), however little of this is exposed to air. The surface area of in-situ snow exposed to the atmosphere is roughly 0.013 m^2/kg

(assuming 1-mm grain diameters). Blowing snow has a surface area of over 60 m^2/kg exposed to the atmosphere (presuming 0.1-mm grain diameters) and is ventilated at an extremely high rate compared to surface snow.

ii) *Blowing snow undergoes sublimation.* Because of rapid energy exchange there is a phase change on the blowing snow particle surface where ice sublimates to water vapour. Intermediate liquid-like phases on the particle surface may be promoted by this transformation. Any solute or particulates in the shrinking ice lattice would be released to this liquid-like layer or possibly the atmosphere.

Snow Accumulation Model

Snowfall = 30 mm
RHmax = 90% for T simulation
Tmax = -15 C for RH simulation

— T = -5
— T = -15
- - T = -25
— RH = 70

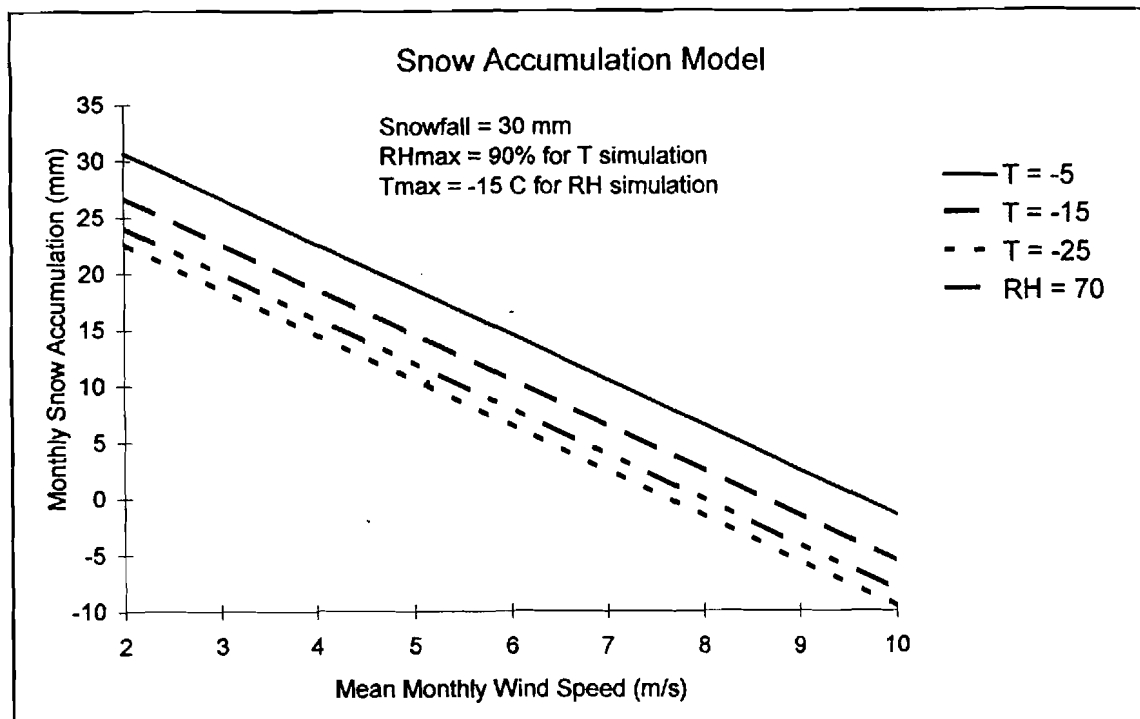Monthly Snow Accumulation (mm)

Mean Monthly Wind Speed (m/s)

Figure 3. Monthly snow accumulation in windswept, open environments as a function of monthly snowfall, mean monthly wind speed and the monthly means of daily maximum air temperature and relative humidity.

Photochemical Reactions

Blowing snow and sublimating surface snow are particularly well-exposed to the atmosphere and to solar radiation and may therefore undergo photochemical reactions. Neubauer and Hermann (1988) suggested that photochemical reactions could cause loss of NO_3^- from surface snow in polar regions. Their hypothesis states that the loss of NO_3^- is due to the re-emission of NO_2 formed from the photodegradation of HNO_3 by solar radiation. The effective range of wavelengths that induce this reaction is approximately 300-320 nm (maximum absorption 302 nm). The reaction can actually form NO_2, the nitrite ion NO_2^-, atomic oxygen and the hydroxyl radical. Zepp et al. (1987) found that the irradiation of NO_3^- in the presence of OH-radical probe-molecules showed that the OH radical produced is a powerful oxidant. Thus the photodegradation of HNO_3 may not only result in loss of NO_3^- but also in the concomitant oxidation of other reduced species e.g. S(IV). Atomic oxygen combines with dioxygen to produce ozone which, with the hydroxyl radical, is an intermediate in the aqueous peroxide ($H_2O_2 - O_2^-$) reaction pathways (Gunz and Hoffman, 1990). It should be noted that these reactions take place in water; because quasi-liquid layers are also produced on snow grain surfaces from heating during solar irradiation then photochemical reactions on snow surfaces are a distinct probability (Jones et al., 1993). Conklin and Bales (1993) showed that a liquid layer thickness of 3-30 nm develops at -60° C increasing to 500-3000 nm at -1° C, so concommitant irradiation and heating are prerequisites for photochemistry on ice crystals. Though blowing snow has a high surface area that can be exposed to the sun, blowing snow particles are in transit for relatively short times (minutes or less) which may be insufficiently long for photochemical reactions to occur. Sublimating surface snow, with associated liquid-like layers on the ice surface, is a better candidate surface for photochemical transformation. The same energy source (radiation) that primarily drives sublimation of surface snow also drives the photochemical processes and warms the upper snowpack; this makes chemical changes resulting from either process difficult to distinguish in the field.

Phoresis

Beard et al. (1983) noted that evaporating drops the size of blowing snow particles (60-90 μm) can scavenge aerosols by thermophoresis, diffusiophoresis and electrostatic attraction. Sublimation creates strong gradients of temperature and water vapour density between the relatively damp, cool air near blowing snow particles and the ambient atmosphere. These gradients will cause thermophoretic and diffusiophoretic tendencies, though the magnitude of any scavenging due to these processes is not known for blowing snow. Miller and Wang (1989) indicated that for falling snow particles in undersaturated air the collection efficiency for aerosols from 0.1 to 1.0 μm in radius increases an order of magnitude as relative humidity decreased from 95% to 50% - such a decrease in humidity would cause a dramatic increase in sublimation rate and in theory thermo- and diffusiophoresis. Murakami et al (1985) and Sauter and Wang (1989) suggest that aerosol collection efficiency decreases with increasing size and compactness of a snow crystal. This is confirmed by field measurements (Lamb et al., 1986) that show small snow particles are more efficient than large particles in phoretic capture of aerosols - small particles also have higher sublimation rates per unit mass.

Electrostatic charge can increase the aerosol collection efficiency of snow crystals by two orders of magnitude, making electrophoresis an extremely important scavenging mechanism (Miller and Wang, 1989). Wishart (1970) measured the electrification of blowing snow in the Antarctic, a phenomenon which has widespread evidence in polar regions by its disruption of electronic communications during blizzards; a typical charge is -50 μC/kg. Pomeroy et al. (1991a) suggested that particle charge might contribute to scavenging of sea-salt aerosols during blowing snow and measured strong charges concurrent with rapid aerosol scavenging by blowing snow. An inverse relationship between ventilation velocity and collection efficiency due to electrophoresis has been measured, suggesting that small blowing snow particles will more effectively collect aerosols by electrophoresis (Murakami and Magono, 1983). Such a hypothesis would support electrophoretic forces being generated by sublimation as noted in the laboratory by Dong and Hallett (1990). Schmidt and Dent (1993) examined electrostatic forces on blowing snow particles and concluded that crystal fragmentation, asymmetric rubbing and transient contact of saltating particles with the surface contribute to a thermoelectric effect which is responsible for charges on blowing snow particles. The thermoelectric effect results from a temperature gradient across an ice crystal

producing an ionic concentration gradient, along which H^+ ions move more rapidly than do OH^- ions (Latham and Mason, 1961). Latham and Mason showed that when two pieces of ice were put in contact and then separated, the warmer of the two developed a negative charge whilst the cooler developed a positive charge. The charge difference relates to the surface layers of the particles as affected by their temperature. A charge structure exists within the quasi-liquid layer surrounding snow particles (Baker and Dash, 1994). During transient contact between two ice particles (or a snow crystal and the snow surface) the quasi-liquid layer will flow from the thicker to the thinner layer and result in a charge transfer. The thermoelectric effect from particle impact implies that electrophoresis will occur most strongly near the surface near the saltating snow layer, however any additional electrophoresis due to sublimation would reach a maximum well above the surface. Resulting electrophoretic scavenging by blowing snow is probably most efficient in the saltation layer but still notable above this layer.

Ventilation

Ventilation of blowing snow particles contributes to scavenging of aerosols by interception and inertial impact and to turbulent diffusion of soluble gases such as HNO_3, H_2O_2, HCl and NH_3 to the snow particle surface (Dawson and Brimblecombe, 1983; Huebert et al., 1983). Concomitant with ventilation is sublimation of ice from the snow particle which may concentrate certain chemicals on the ice crystal surface and release certain chemical species from the sublimating particle (Delmas and Jones, 1987). Snow particles may be blown for distances of 3 to 5 km in temperate and low Arctic conditions (Tabler et al., 1990a) increasing to tens of kilometres in cold polar conditions (e.g. at -40 $^\circ$C). Pomeroy et al. (1991a) provided an expression for the ventilation velocity, v_r (m/s), of blowing snow particles that includes turbulent and average components as a function of particle radius, r (m), and ambient mean wind speed, u_z (m/s), where

$$v_r = 1.1 \times 10^7 \ r^{1.8} + 0.01061 \ u_z^{1.36} \qquad (9)$$

Typical mean particle radii in the saltation layer are 100 μm and at 2-m height in the suspended layer are 40 μm. Given a typical particle radius of 50 μm and ambient wind speed

of 8 m/s the particle ventilation velocity would be 0.38 m/s, considerably in excess of that due to wind pumping or wind shear on surface snow.

Huebert et al. (1983) has modelled and Zinder et al. (1988) have measured gaseous scavenging of HNO_3 for falling snow - the result should be applicable to blowing snow when sublimation rates are very small (e.g. humid conditions). Zinder et al found that concentrations of NO_3^- in falling snow particles increased 50% over a 1200m fall in a humid atmosphere.

Where ventilation is accompanied by rapid sublimation then high concentrations of certain chemical species will result on the surface of a snow particle. These concentrations may exceed the solubility of these species in the liquid-like surface layer and result in volatilisation. This is particularly expected for volatile species such as HNO_3 and H_2O_2 whose rate of volatilisation may be augmented by the increase in acidity due to higher concentrations of H_2SO_4 and other non-volatile acids in the liquid-like layer. It is also likely that H^+ ions will migrate to the cool surface of a sublimating snow particle, causing further ionic imbalance and potential for volatilisation. The net effect of ventilation may be volatilisation when sublimation rates are high, but in instances of high atmospheric concentrations and low sublimation rates then gaseous scavenging by the blowing snow particle will result.

Chemical Deposition in Wind-blown Snow

The concentration change due to blowing snow may be found by a mass balance of solute and solvent, where CF_x, a dimensionless concentration factor, is the concentration of any species x, at time t, normalised to its concentration at initial time i:

$$CF_x = \frac{[\frac{m_x(i) + m_x(s) - m_x(v)}{m_m(i) - m_m(E)}]}{[\frac{m_x(i)}{m_m(i)}]} \quad (10)$$

m is the load of the chemical species (m_x, $\mu eq/m^2$) or snow (m_m, mm water equivalent) and s refers to scavenging gains, v to volatilisation losses and E to sublimation losses. Loss by

chemical transformation of species is assumed to be negligible. Equations 6, 7 and 8 can be used to evaluate snow mass changes where meteorological information is incomplete. Recognising that there are potential errors which accrue in averaging snow chemistry over one month, a simple model for monthly concentration of accumulating snow, C_M (µeq/l), is

$$C_M = \frac{M_x(i) + M_x(s) - M_x(v)}{M_m(i) - M_m(E)}$$

(11)

where M_x is a monthly load of chemical species in snow (µeq/(m^2 month)), M_m is a monthly snow flux (mm/month) and (i) refers to parent precipitation, (s) refers to monthly scavenging, (v) refers to monthly volatilisation and (E) to monthly sublimation.

The monthly derivation of concentration allows Eq. 10 to be recast in a monthly version,

$$CF_m = \frac{1 + \dfrac{M_x(s)}{M_x(i)} - \dfrac{M_x(v)}{M_x(i)}}{1 - \dfrac{M_m(E)}{M_m(i)}}$$

(12)

where CF_M is the monthly average concentration factor. Scavenging or volatilisation must be inferred from empirical relationships, modelled results or the results of field campaigns. Equation 11 may be adapted to determine monthly chemical species accumulation or monthly load, M_x, by reference to Eq. 5 as,

$$M_x = C_M [M_m(i) - M_m(T) - M_m(E)]$$

$$and$$

(13)

$$M_x = [M_x(i) + M_x(s) - M_x(v)] (1 - \frac{M_m(T)}{M_m(i) - M_m(E)})$$

This mass balance calculates $dQ_T/dx = -M_m(T)$. Equation 13 embodies a mass balance where a quantity of snow is removed ($M_m(T)$) and the concentration of this snow is determined by $[M_m(i) - M_m(E)]$. Hence, sublimation plays a role in the accumulation of chemical species

by controlling the concentration of ion in snow that is removed from the site by wind transport.

Presuming for the purpose of example that $M_X(s)$ and $M_X(v)$ are negligible, the effects of varying wind speed, temperature, humidity and fetch on the concentration factor, CF_m, and the net ionic load, $M_X/M_X(i)$, are shown in Fig. 4. Note that net ionic load is not equivalent to concentration as the snow removed may have a different chemical concentration than the parent or final accumulating snow. The model shows that concentration factors due to blowing snow increase sharply as sublimation increases with wind speed and less dramatically with decreasing temperature and relative humidity. CF_m can range from 1.5 to 2 for short fetches and up to 2 to 6 for longer fetches in the conditions typically found in polar regions. The net ionic load, $M_X/M_X(i)$. decreases rapidly with increasing transport and sublimation fluxes and is hence strongly related to wind speed, reaching 0 at wind speeds above 8 to 10 m/s. It decreases more rapidly with lower temperatures but is relatively insensitive to humidity. Because blowing snow sublimation increases with fetch length, the net ionic load for longer fetches remains near 1, until the wind speed is reached at which all snow is removed from the site by erosion. The transition between these states is quite dramatic and is reached at a wind speed between 8 and 10 m/s. For the longer fetches typical of polar ice caps Eq. 13 has important implications because an almost binary system develops in which relatively complete ionic load (though highly concentrated) persists for low wind speeds switching rapidly to almost no ionic load for higher wind speeds. This result is not unexpected considering Fujii's (1981) observation that the formation of an annual layer of snow accumulation occurs only once every two or three years because of sublimation and wind erosion of snow. As the transition wind speed between the two states is near to the long-term monthly average at many Antarctic stations this aspect of wind-blown snow chemical accumulation makes the process highly non-linear and sensitive to atmospheric conditions (wind speed and temperature in particular). One should note that the previous simulation presumed that all species were perfectly conserved in sublimating snow particles, subsequent evidence will show that this is not the case. For species that volatise at the same rate that ice sublimates there would be little change in concentration but a dramatic drop in load as wind speed increases.

Blowing Snow Concentration Factor

Concentration Factor

3.5

3

2.5

2

1.5

1

T = -5   Fetch = 1 km

T = -15

T = -25

RH = 70%

2   4   6   8   10

Wind Speed (m/s)

Net Ionic Load of Windblown Snow

Net Ionic Load (M/Mi)

1

0.8

0.6

0.4

0.2

0

Fetch = 1 km

T = -5

T = -15

T = -25

RH = 70%

2   4   6   8   10

Wind Speed (m/s)

Blowing Snow Concentration Factor

Concentration Factor

10
9
8
7
6
5
4
3
2
1

Fetch = 3 km

T = -5

T = -15

T = -25

RH = 70%

2   4   6   8   10

Wind Speed (m/s)

Net Ionic Load of Windblown Snow

Net Ionic Load (M/Mi)

1

0.8

0.6

0.4

0.2

0

Fetch = 3 km

T = -5

T = -15

T = -25

RH = 70%

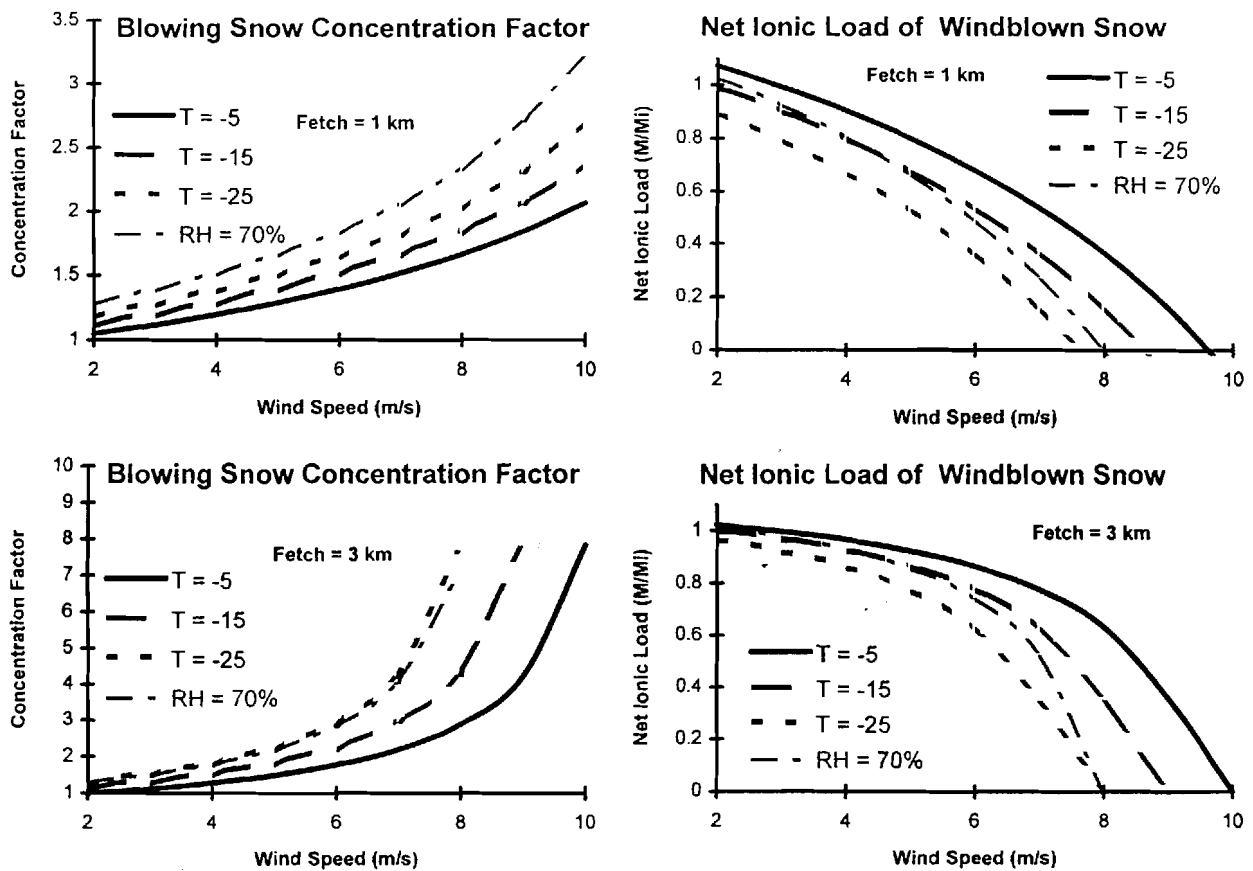2   4   6   8   10

Wind Speed (m/s)

Figure 4. Effects of wind speed and atmospheric environment on Concentration Factor (CF) and ion load for wind-blown snow. All simulations are for RH_{max} = 90%, except for that at 70% for which T_{max} = -15°C.

EXPERIMENTAL EVIDENCE

Experimental evidence for changes to the chemistry of wind-blown snow exists for a few locations around the world; the small number of studies is more a reflection of the difficulty of obtaining robust field data in extreme weather conditions than lack of interest by the scientific community.

Osada (1994) measured the concentration of several major ions (Cl^-, NO_3^-, SO_4^{2-} and Na^+) in blowing snow under katabatic conditions near Mizuho Station in East Antarctica. Unfortunately the concentration of ions in falling snow was not reported for comparison. A marked seasonal change in concentration for NO_3^- and excess (to sea-salt) SO_4^{2-} was found with mean concentrations in the austral summer of 4.63 and 3.10 μeq/l respectively and in the austral winter of 0.97 and 0.37 μeq/l. A dramatic order of magnitude change in the ranges of concentrations occurs over a few weeks in March and November. Osada notes that the seasonal variation shows some correspondence with changes in the concentration of atmospheric aerosols, however the details of the variation do not match. Blowing snow concentrations show sharper rates of seasonal change than do aerosols and no evidence of the mid-winter peak of NO_3^- in aerosols. The author suggests that differences in the transport of aerosols and acidic gases or rapid scavenging of aerosols by blowing snow might account for the variation in concentration, however sublimation of blowing snow and volatilisation of chemical species were not considered.

Mulvaney and Wolff (1994) survey the spatial distribution of major ions in the Antarctic ice sheet. There is a roughly 10-fold decline in sea-salt ions as represented by Cl^- with distance from the coast; this represents a scavenging loss as the aerosols move inland from the ocean. Excess SO_4^{2-} sometimes decreases with distance from the coast (two-fold) and at other locations does not. The authors suggest that the scavenging mechanism is more efficient for sea-salts than for excess SO_4^{2-}. Of interest is a three-fold increase in SO_4^{2-} concentrations as one goes furthest inland (East Antarctica) into regions of low precipitation and high wind speeds. NO_3^- concentrations are relatively uniform and show little trend with distance from the coast but tend to increase with elevation (three-fold) and decrease with accumulation rate (three-fold). An additional seasonal variation in NO_3^- has been measured and though attributed to variable sources, post-depositional loss has not been ruled out (Mulvaney and Wolff, 1993). The spatial variation of chemical concentrations are attributed

to various processes by Mulvaney and Wolff (1994), but may additionally be explained by blowing snow processes. For example, the decline of sea-salt ions from the coast is due to scavenging, part of this scavenging could be due to electrophoretic attraction to blowing snow particles. Similarly, near the coast, excess SO_4^{2-} is scavenged but not as efficiently as the sea-salt species; this is consistent with blowing snow scavenging. The increase in excess SO_4^{2-} further inland can be explained by the higher CF values due to relatively large sublimation in that low precipitation zone. The uniformity of NO_3^- concentrations is consistent with volatilisation of HNO_3 during sublimation. However the moderate increase with elevation and decrease with precipitation indicates the species does not completely volatise during sublimation.

Delmas and Jones (1987) measured the concentrations of SO_4^{2-}, NO_3^- and NH_4^+ in wind-blown and parent snow on a frozen lake in the boreal forest of Québec. Initial concentrations were 107.8, 9.5 and 1.9 µeq/l for NO_3^-, SO_4^{2-} and NH_4^+ respectively. After one day of blowing snow without precipitation the concentrations dropped 23% for NO_3^-, 20% for SO_4^{2-} and 14% for NH_4^+. The wind speeds were at the threshold for blowing snow and Québec winters are relatively humid so the sublimation rate may have been small in this case. Interestingly, atmospheric concentrations of N and S species were high during the loss from the snowpack. Delmas and Jones suggest that the decrease is due to resuspension of aerosols from the saltating snow particles. Saltating particles are abraded and in this case would undergo a small degree of sublimation which might promote detachment of aerosols from the snow crystal.

Pomeroy et al. (1991a, b) measured the chemistry of fresh, blowing and wind-blown snow on a highland plateau in the Cairngorm Mountains, Scotland. The location is in a relatively temperate environment, however extremely high wind speeds, frequent snowfalls and highly-concentrated acidic contaminants in snow, permit an unequivocal interpretation of blowing snow chemistry processes. Three types of snow were collected during a blowing snow event without concurrent precipitation and analysed for the major anions and cations: fresh undrifted snow, blowing snow at several heights above the surface and wind-blown snow that had undergone relocation by the wind. The sequence of ion concentrations from undrifted to blowing snow to wind-blown snow is shown in Fig. 5. All ion concentrations in blowing snow were higher than the parent snow concentrations, as much as three times higher in some cases (Cl^-, Na^+, K^+). Ion concentrations of Cl^-, SO_4^{2-}, Na^+, K^+ and Mg^{2+} were
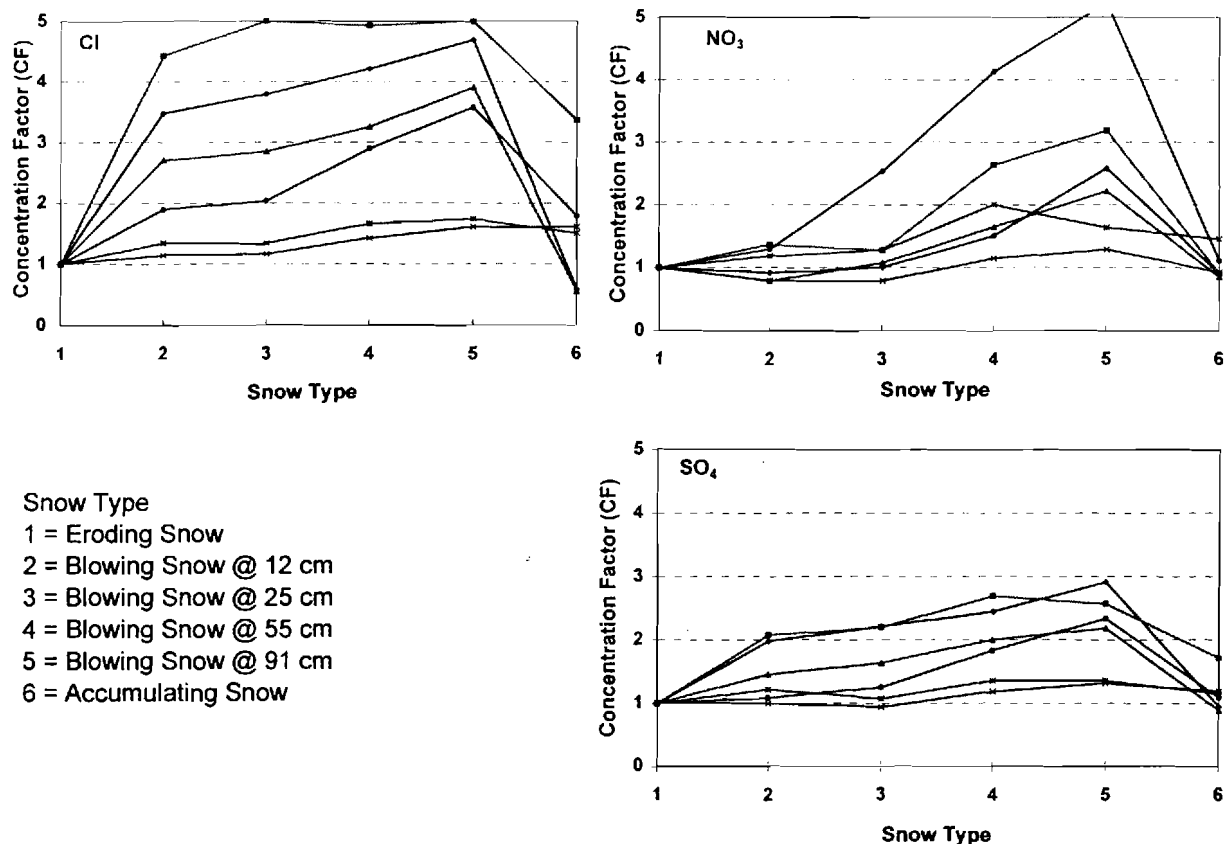
Figure 5. Sequence of ion concentration from undrifted to blowing snow to wind-blown snow as measured in the Cairngorm Mountains, Scotland (after Pomeroy et al., 1991).

significantly higher in wind-blown snow than in the parent snow (80% confidence), however NO_3^- showed little difference between the two. Hence, the elevated concentrations in blowing snow are reflected in wind-blown snow for all ions except for NO_3^-. Wind-blown snow ion concentrations are generally less than blowing snow concentrations because the wind-blown snow was a mixture of snow that had travelled at great heights (and therefore distance) with resultant high concentrations and creeping and saltating snow, which, though relocated, possessed relatively lower concentrations. A combination of ice sublimation, species volatilisation and particulate scavenging processes was suggested to account for the observed increase in concentration during wind transport. Pomeroy et al. (1991a) calculated the concentration factor due to sublimation using meteorological measurements and a range of hypothetical particle trajectories; the concentrations of SO_4^{2-}, NO_3^- and NH_4^+ in blowing

snow matched that expected from the parent snow and the concentration factor (CF), however Na^+, K^+, Mg^{2+} and Cl^- concentrations were two-fold in excess of that expected from the CF due to sublimation. This excess concentration to the CF is most pronounced for blowing snow derived from the saltation layer and therefore may be due to electrophoresis; the only known scavenging process at a maximum in the saltation layer. The apparent loss of NO_3^- from wind-blown snow subsequent to deposition was dramatic and suggests a consumption or volatilisation as HNO_3. It is possible that before wind transport occurred, concentrations of NO_3^- may reflect an equilibrium between that on the ice surface and HNO_3 in the atmosphere. After rapid sublimation the NO_3^- ions are relatively concentrated on the ice particle surface and possibly in solution as HNO_3; when subject to further but much slower ventilation due to wind pumping (see Waddington this volume) the concentrations on the ice surface readjust to reach an equilibrium with the partial pressure of HNO_3 in the atmosphere via a vapour transfer from particle surface to atmosphere. Increased acidity from high H_2SO_4 concentrations on the sublimated ice crystal may further promote such a transfer. The result is a loss of NO_3^- in wind-blown snow that is roughly proportional to the loss of ice from the previously sublimating blowing snow particle.

Field evidence of the chemical transformations caused by wind-blown snow exists for the Canadian Western Arctic; a region remote from most ionic sources (except sea-salt) and where a strong gradient in annual blowing snow frequency exists over several hundred kilometres (Pomeroy et al., 1995). In the southern tundra-taiga transition (south of Inuvik, NWT) blowing snow is infrequent and concentrations of the three major ions in snow are relatively low ($Cl^- = 6.2$, $NO_3^- = 2.6$, $SO_4^{2-} = 4.5$ µeq/l). In the open tundra (Tuktoyaktuk), blowing snow causes 58% of annual snowfall to sublimate before accumulation and resulting ion concentrations are much higher ($Cl^- = 47.1$, $NO_3^- = 4.5$, $SO_4^{2-} = 10.8$ µeq/l). Near Inuvik and straddling the transition is Trail Valley Creek where 31% of snowfall sublimates as blowing snow. The CF due to sublimation and the measured CF for the three ions are shown in Table 1. Data derive from landscape stratified surveys with coefficients of variation for concentrations and snow water equivalent being quite consistent and increasing from 0.05 for NO_3^- south of Inuvik to 0.21 for Cl^- at Tuktoyaktuk. The higher Cl^- concentrations (7.6 times higher) are certainly related to Tuktoyaktuk's proximity to the Arctic Ocean. The highest Cl^- concentrations were found where the snow had undergone the most drifting, suggesting additional enhancement by electrophoretic scavenging and sublimation. The CF

values due to blowing snow sublimation closely match those for SO_4^{2-} and overestimate those for NO_3^- in the transition from a non-transport to severe blowing snow regime. This is further illustrated by the SO_4^{2-}/NO_3^- ratios which increase with frequency of blowing snow from 1.73 south of Inuvik, to 1.94 at Trail Valley and to 2.4 at Tuktoyaktuk. These results concur with the result from Pomeroy et al. (1991a, b) that concentrations with respect to blowing snow sublimation of SO_4^{2-} are conservative, Cl^- are enhanced and NO_3^- are not fully retained in wind-blown snow.

Table 1. Concentration Factors (CF) Measured, Spring 1993 and due to Blowing Snow Sublimation, Winter 1992-93 - Canadian Western Arctic.

Site	Landscape	Sublimation	CF Subl.	CF Cl^-	CF NO_3^-	CF SO_4^{2-}
South Inuvik	Taiga	0 %	1.0	1.0	1.0	1.0
Trail Valley	Transition	31%	1.45	2.8	1.3	1.5
Tuktoyaktuk	Tundra	58%	2.38	7.6	1.7	2.4

Further field evidence from the same region shows conflicting evidence for ion conservation during sublimation. Jones et al (1993) placed weighed, homogenised snow of known chemistry in mesh bags and exposed them on open snow surfaces to wind and either sunlight or shade for 36-48 hours in late winter 1992. One set of bags was placed near Inuvik (temperature -8 to -10 ^{o}C and low humidity) and one set of bags placed in Trail Valley (temperature -5 to -15 ^{o}C and higher humidity). The proposed test of photochemical reactions indicated that changes in chemical composition after the exposure were not attributable to exposure to sunlight or shade. Of interest, however was a steady, slow sublimation from snow in the mesh bags at the Inuvik site. The bags in the warmer, low humidity environment underwent sublimation and lost on average 50 g or about 11-22% of their mass of snow during this period. The bags in the colder, high humidity environment underwent no sublimation and lost no mass. For the sublimating bags there was no statistically significant dry deposition of either NO_3^- or SO_4^{2-}, nor was there volatilisation - each species was completely conservative. For the non-sublimating bags there was no exchange of NO_3^-, but a dry deposition of SO_4^{2-} equivalent to 2.5 $\mu eq/(m^2$ day).

Sublimation of 11-22% over several days is slow compared to that which occurs from blowing snow particles; in this "slow sublimation" case there is complete retention of NO_3^- by the sublimating snow particles and no dry deposition.

Recently, a cold room laboratory experiment was used to test the effect of in-situ snow sublimation on chemical concentrations in snow under a controlled condition. A homogenised column of natural, cold prairie snow was placed in a climatically and atmospherically controlled cold box and maintained at $-4^{\circ}C$, 70% relative humidity with an atmosphere of medical grade air. A bright beam of light, spectrally controlled to match the intensity and spectrum of sunlight in polar regions was shown continuously for 3 to 4 days onto part of the snow column. Ion concentrations in irradiated surface snow are similar to those in non-irradiated surface snow, hence no photochemical effects were evident in this experiment. However, sublimation of surface snow layers was caused by the radiative energy input to the snow. In the first experiment run, parent concentrations of NO_3^- and SO_4^{2-} were 10.2 and 9.4 µeq/l and CF of 1.14 and 1.36 respectively were measured after the sublimation treatment. This changed the SO_4^{2-}/NO_3^- ratio from 0.92 in parent snow to 1.10 in the sublimated snow. For the second experiment run a fan was operated to recirculate air within the chamber and increase the sublimation rate. Parent concentrations of NO_3^-, SO_4^{2-} and NH_4^+ were 9.3, 5.2 and 4.2 µeq/l and CF of 1.47, 1.88 and 1.96 were measured after the sublimation treatment. In this case the SO_4^{2-}/NO_3^- ratio increased from 0.56 in the parent snow to 0.72 in the sublimated snow. Mass losses from sublimating snow in weighed petri dishes in the chamber confirm that SO_4^{2-} and NH_4^+ were relatively conservative during sublimation but NO_3^- was lost. For the two experimental runs from 16% to 22% of NO_3^- was lost from the surface layers when sublimation removed from 1/4 to 1/2 of the snow in these layers.

IMPLICATIONS FOR POLAR REGIONS

The experimental evidence suggests that notable and complex changes occur to chemical concentrations as snow is relocated. There are five categories into which the changes can be classified, with respect to the transformation of chemical species during blowing snow:

i. *Conserved* - retained by the snow crystal during sublimation, low scavenging rates and little resuspension during transport (e.g. SO_4^{2-}),

ii *Scavenged* - retained by the snow crystal during sublimation, high scavenging rates due to primarily phoretic effects and secondarily gaseous uptake and little aerosol resuspension during transport (e.g. Cl^-, Na^+, Mg^{2+}, K^+),

iii *Volatised* - lost via vapour transfer from the snow crystal during sublimation (possibly NO_3^-),

iv *Resuspended* - lost via aerosol resuspension from the snow crystal during sublimation and abrasion (possibly NO_3^-, SO_4^{2-} and NH_4^+),

v *Consumed* - consumed by reaction within the snow crystal during blowing snow or after deposition (no confirmed examples, possible photochemical reactions, though these are usually assumed not to occur or are included in the volatilisation term).

The complex physio-chemical nature of the phenomenon means that no ion can be strictly categorised; an ion may display the tendencies of one or more categories at one time and change its behaviour under different environmental conditions. For instance NO_3^- has been observed to be conserved at low sublimation rate conditions (usually in-situ), volatised as HNO_3 during rapid snow sublimation (during blowing snow) and resuspended as an anthropogenically-derived particulate under blowing snow with low sublimation rates. The dual nature of NO_3 as part of a gas or of an aerosol, variable partial pressures, its location on the ice surface and possible particle surface reactions probably play a role in this complex behaviour. The normal conservation of SO_4^{2-} but occasional resuspension as an aerosol will relate to the position of the aerosol on the ice crystal, the depth of any quasi-liquid layer and the rate of sublimation. In most studies SO_4^{2-} is found to be completely conservative but Delmas and Jones (1987) showed that snow crystals that undergo shattering in saltation transport and little sublimation appear to resuspend anthropogenic SO_4^{2-}. When considering the accumulation in glacial snow, the behaviour of an ion may depend upon the severity of blowing snow sublimation. Following the model presented in Eq. 8 and Fig. 3 a binary system is proposed. For wind transport conditions when up to half the annual snowfall sublimates each year then many ions are either conserved or scavenged and likely undergo a

concentrating effect after each blowing snow sublimation event, becoming more concentrated in accumulating snow. This corresponds to the lower wind speed condition in Fig. 3. NO_3^- is the known exception to this trend, being volatised even under relatively moderate blowing snow regimes. When blowing snow removes a large portion of the annual snowfall in several severe blizzards, then many species may be resuspended and volatised along with the remnants of the initial snow crystals. This corresponds to the higher wind speed condition in Fig. 3. These ions will then contribute little to the accumulation layer. As the transport and sublimation rates of blowing snow increase exponentially with wind speed and the wind speed undergoes seasonal and annual trends, it is quite possible that polar environments experience both types of transformation and substantial variation in the dominant mode of transformation seasonally, annually and historically.

To examine blowing snow regimes in polar environments and compare them to more intensively studied continental environments, a special run of the Hadley Centre (UK) GCM (1994 version) was used to develop a spatially-precise climatological database for the central Antarctic and central Greenland. The GCM was run for 10 years as part of a broader applications study to provide averages of *present* conditions in these remote locations. This method avoids use of wind speed data from automated weather stations which are subject to severe riming in mid-winter and snowfall measurements that are subject to undermeasurement and hence induce large errors in calculating a blowing snow climatology. Unfortunately the GCM appears to overestimate humidity in most winter conditions and underestimate winter precipitation in polar regions as it estimates accumulation not snowfall (Connolley and Cattle, 1994). Table 2 shows some average meteorological conditions in January and July as predicted by the GCM in four selected locations, central Greenland, the Canadian Prairies, coastal Antarctic and central Antarctica. The meteorological variables shown are important in estimating the monthly blowing snow fluxes using Eq. 7.

Table 2. Climatology of three blowing snow regimes (north polar, continental, south polar) as estimated from the Hadley Centre GCM.

Location	January Max. Daily Temperature	July Max. Daily Temperature	Mean Monthly Wind Speed m/s
Central Greenland 75°18'N 40°45'W	-41 °C	-11 °C	5.9
Canadian Prairies 52°38'N 108°30'W	-8 °C	24 °C	4.7
Central Antarctica 82°05'S 33°22'E	-27 °C	-69 °C	5.1
Coastal Antarctica 74°49'S 82°15'W	-7 °C	-23 °C	7.0

Annual blowing snow sublimation calculated using Eq. 7 and the GCM simulation is shown in Table 3 for the four locations, along with snowfall and the CF due to blowing snow sublimation, presuming a perfectly conservative ionic species. The CF values must be considered crude, first approximations. Sublimation is calculated for two fetch lengths; one kilometre and ten kilometres. Equation 7 provides the monthly sublimation fluxes for a 1 km fetch typical of mixed agricultural landscapes or irregular terrain. This 1-km sublimation is adjusted for the 10-km fetch more appropriate to the polar ice caps by presuming that all transported snow (1-km) except the 10% required to maintain a mass balance has sublimated over the 10-km fetch, and adding this quantity to the 1-km sublimation. Sublimation is underestimated and precipitation overestimated for the Canadian Prairies; actual values are roughly 110 mm snowfall and 40 mm sublimation (Pomeroy and Gray, 1995). The 10-km sublimation values exceed the GCM-modelled snowfall in both central Antarctica and Greenland; this is likely due to an underestimation of snowfall or an overestimation of sublimation. There has been no verification of the blowing snow sublimation routine poleward of 70°N and it may overestimate sublimation in persistently cold conditions; however, as the GCM has been calibrated against polar *accumulation* rather than actual precipitation, the snowfall underestimation is likely to be quite large. What the modelling exercise does clearly

indicate is the substantial variation possible in CF for conservative species within the present range of polar climates and the difficulty in determining the exact CF with present meteorological data sets and knowledge of polar blowing snow. For example, modelled blowing snow in central Antarctica removes most snow and ions, but in coastal Antarctica ions are deposited in a more concentrated state. In central Greenland, the modelled blowing snow regime borders between removal and concentration states, depending upon the fetch distance and probably varies in state from year to year. In all polar locations the state varies seasonally. A full-scale field investigation of the transformations of various chemical species during blowing snow in polar atmospheric conditions is required to begin to predict actual CF values in relation to that of a perfectly conserved species, to predict the onset of resuspension, and to model the transformations of volatile and scavenging species.

Table 3. Annual blowing snow sublimation for three regimes (north polar, continental, south polar) as estimated using the blowing snow model and data from the Hadley Centre GCM. All values are annual estimates. *** denotes CF impossible to calculate but estimated as quite large.

Location	Snowfall	Blowing Snow Sublimation 1-km Fetch	Blowing Snow Sublimation 10-km Fetch	CF 1 km	CF 10 km
Central Greenland	151 mm	85 mm	218 mm	2.28	***
Canadian Prairies	151 mm	12 mm	30 mm	1.09	1.25
Central Antarctica	48 mm	73 mm	242.2 mm	***	***
Coastal Antarctica	627 mm	115 mm	244 mm	1.22	1.64

SUMMARY

Blowing snow is ubiquitous in polar regions and undergoes relatively rapid sublimation whilst in transit. The erosion, transport and deposition of blowing snow dramatically transforms the physical and chemical characteristics of polar snow. Processes active during snow transport are sublimation, thermophoresis, diffusiophoresis, electrophoresis and ventilation; these processes occur at much higher rates than for "in-situ" surface snow. Conditions suitable for photochemistry exist on blowing snow particles though no strong empirical evidence exists for such reactions in snow. Annual accumulations of snow and ions on polar ice-sheets are a residual of the blowing snow phenomenon and therefore a result of the interaction of wind speed, temperature and snowfall regimes with surface topography and the meteorological history of the snowpack. Chemical transformations in blowing snow involve ion conservation, scavenging, volatilisation, resuspension and consumption. Experimental evidence from Canada, Scotland and the Antarctic shows that for moderate blowing snow regimes the first three processes dominate whilst for severe regimes resuspension dominates. In moderate blowing snow, concentration increases are greatest for scavenged ions such as those of sea-salt aerosols. Concentration increases are proportional to sublimation loss for conservative species such as SO_4^{2-} and NH_4^+ and less than proportional to the sublimation loss for volatile species such as HNO_3. Concentration increases due to sublimation increase exponentially with wind speed and vary widely with climate and terrain in polar regions. Annual ion load depends on the blowing snow regime and displays two almost binary modes of retention and resuspension. For polar atmospheres a wind speed of 8 m/s, near the mean for many ice sheets, divides the two depositional modes. Hence the process of ion deposition in snow in polar regions is inherently unstable and varies in mode seasonally and annually. The uncertainty in estimating and spatially extrapolating wind speed regimes over polar regions suggests that ion concentration factors and annual retention of ions in wind-blown snow are probably unpredictable over even recent history. This adds considerable complexity to attempts to interpret the mass balance and chemistry of recent ice cores and link this interpretation to atmospheric chemistry.

CONCLUSIONS

1. Blowing snow is ubiquitous in polar regions and dramatically transforms the physical and chemical characteristics of polar snowcovers. Associated with transport of snow particles are the processes of sublimation, thermophoresis, diffusiophoresis, electrophoresis and ventilation which occur at much higher rates than for in-situ surface snow. Conditions suitable for photochemistry may occur on sublimating snow particles.

2. Accumulation of snow and constituent ions in polar regions is a residual of blowing snow fluxes and therefore a function of the wind speed, temperature and snowfall regimes. Sublimation fluxes in polar regions exceed accumulation fluxes by several fold and approach "true" snowfall fluxes in extreme environments.

3. Transformations of blowing snow chemistry may involve species conservation, scavenging, volatilisation, resuspension and consumption. For moderate blowing snow regimes the first three transformations dominate, whilst for severe regimes resuspension dominates. Consumption is undocumented.

4. Conservative species are exemplified by SO_4^{2-} and NH_4^+, scavenging species by those of sea-salt aerosols and volatising species by HNO_3.

5. For conservative species a concentration increase due to sublimation occurs in polar regions; the increase is quite sensitive to blowing snow regime, increasing exponentially with wind speed and varying widely with climate and terrain.

6. Snow and ion loading are dependent upon the blowing snow regime and show an almost binary state of retention and resuspension. For polar atmospheres the boundary between retention and resuspension occurs at mean wind speeds of about 8 m/s above which relatively little snow or ions are retained on the ice caps. This wind speed is near the monthly averages for many interior Antarctic and central Greenland locations, suggesting that the ion deposition process in the Antarctic and Greenland is inherently unstable and varies in mode seasonally

and from year to year. The sensitivity to local climatic conditions means that even slight changes in micro-climate can cause dramatic changes in the annual deposition of chemical species in polar snow.

7. The sensitivity of blowing snow to the variable wind speed regimes over polar regions suggests that ion concentration factors and annual retention of wind-blown snow are probably unpredictable over even recent history. The variable conservation of chemical species by blowing snow is due to combinations of in-transit processes and adds another level of complexity to the system. The dependence of snow ion accumulation processes on synoptic and site specific physical factors and ice-surface chemical transformations adds considerably uncertainty to attempts to interpret the snow accumulation and snow chemistry of recent ice cores strictly in terms of atmospheric chemistry and precipitation.

ACKNOWLEDGEMENTS

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