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SNOWPACK RELEASE OF POLLUTANTS
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MANAGEMENT PERSPECTIVE

This manuscript was prepared under invitation by the Dr. D. C. Adriano, Coordinating Editor of the Springer-Verlag series Advances in Environmental Science. Five volumes of this series are being devoted to scientific reviews of topics on acid rain; this particular contribution (after review) will appear in Volume 2. The paper concisely reviews the current knowlege on snowpack accumulation and losses of pollutants and their effect on the aquatic environment. A major component of NWRI's LRTAP research over the past 7 years has dealt with this important topic, and the invitation to prepare this chapter is a recognition of the quality of our work.

The manuscript contributes directly to the integration and summarization effort now underway in preparation of the inclusive assessment of LRTAP occurrence and effects in Canada that is to be completed by March 1990. In this respect it constitutes a major product of the long-term snowmelt research conducted at NWRI. As it stands, the manuscript bears no direct implications for departmental policies or priorities, but in the end will no doubt influence the 1990 assessment and its forth-coming recommendations.

PERSPECTIVES- GESTION

Le présent manuscrit a été rédigé sur invitation par le D^r D.C. Adriano, Coordonnateur de l'édition de la série Springer-Verlag Advances in Environmental Science. Cinq volumes de cette série sont consacrés à des comptes rendus scientifiques de sujets en rapport avec les pluies acides; la présente contribution paraîtra (après révision) dans le Volume 2. Cet exposé examine de façon concise les connaissances actuelles sur l'accumulation et les pertes des polluants du manteau nival et sur leurs effets sur l'environnement aquatique. Une partie importante du programme de recherche sur le TADPA de l'INRE a porté sur ce sujet au cours des 7 dernières années, et l'invitation à rédiger ce chapitre est une reconnaissance de la qualité de notre travail.

Ce manuscrit a contribué directement à l'effort d'intégration et de synthèse présentement en cours en préparation de l'évaluation des cas de TADPA et de leurs effets au Canada, qui doit être achevée en mars 1990. Il s'agit du fruit des recherches à long terme sur le manteau nival effectuées à l'INRE. Tel qu'il a été rédigé, le manuscrit n'a pas d'implications directes sur les politiques ou les priorités ministérielles, mais il influencera sans doute l'évaluation de 1990 et les recommandations.

ABSTRACT

Information on the snowpack content of major ions, trace metals, and organic contaminants (pesticides and total PCB's) has been reviewed and discussed. While several limitations exist, regional snowpack surveys have been successfully used to delineate spatial trends in acidic deposition. Pollutant concentrations in urban areas are much greater than at regionally representative sites. In contrast to the annual anionic predominance of SO_4^{2-} in atmospheric deposition, NO_3^- is often of a similar magnitude or even greater than SO_4^{2-} in the snowpack. Metal concentrations are generally >10-fold higher at European and North American regional locations than in Arctic or Antarctic "background" sites. The dry deposited component of the total snowpack pollutant load is generally less significant (1-45%) than the wet deposited component although there is much variability among chemical parameters and locations. There is conflicting evidence on the pre-melt stability of snowpacks; stability is clearly governed by many factors and the occurrence of unfrozen underlying soils (thereby allowing a convective heat flux) may be very important. The net radiative energy flux is primarily responsible for melting. The delivery of meltwater is often greatly complicated by the snowpack mesostructure; ice layers and the development of within-pack pipe flow can make modelling of this process very difficult. Rain-on-snow events can be very important both hydrologically and chemically. Fractionation of the pollutants during normal snowpack metamorphosis gives early meltwater ion and metal concentrations that are 5- to 10-fold greater than those in the parent snow. Major ions are not removed from the snowpack at the same rate during melting, a process known as "preferential elution". Surface water chemistry during snowmelt is influenced primarily by the differential loss of snowpack pollutants and meltwater flowpaths. Reduction in pH, ANC, and base cations occur, while Al and NO_3^- levels often increase although the behavior of this last ion is variable from location to location. The effect of snowmelt is generally limited to a near surface layer (1-3 m thick) in lakes; however this is strongly influenced by several physical factors. The spring melt event may hold grave consequences for several species of aquatic biota.

RÉSUMÉ

L'information sur la teneur en ions principaux, en métaux à l'état de traces et en contaminants organiques (pesticides et PCB totaux) du manteau nival a été examinée et analysée. Malgré d'importantes limitations, les relevés régionaux du manteau nival ont été utilisés avec succès pour mettre en évidence des tendances spatiales de dépôts acides. Les concentrations de polluants dans les régions urbaines sont très supérieures à celles qu'on trouve dans des sites représentatifs des régions. Par rapport à la prédominance anionique annuelle de SO_4^{2-} dans les dépôts atmosphériques, le NO_3^- est souvent aussi abondant, si ce n'est plus, que le SO_4^{2-} dans le manteau nival. Les concentrations de métaux sont généralement plus de dix fois supérieures dans certaines zones de l'Europe et de l'Amérique du Nord à celles des zones de "concentrations naturelles" de l'Arctique ou de l'Antarctique. La proportion de la charge des polluants du manteau nival total déposée à sec est généralement moins importante (1-45 %) que la proportion déposée à l'état humide, bien qu'on note une forte variabilité si l'on compare les paramètres chimiques et les emplacements. Les observations concernant la stabilité d'avant fusion des manteaux niveaux semblent être contradictoires; la stabilité est régie de façon évidente par divers facteurs et les cas de sols sous-jacents non gelés (qui permettent un flux de chaleur convective) peuvent être très importants.

Le flux d'énergie radiative est principalement responsable de la fusion. La production d'eau de fusion est souvent fortement compliquée par la mésostructure du manteau nival; les couches de glace et le développement de canaux d'écoulement à l'intérieur du manteau peuvent rendre la modélisation de ce processus très difficile. Les événements de pluie peuvent avoir des effets très importants sur la neige du point de vue hydrologique et chimique. Le fractionnement des polluants au cours de la métamorphose normale du manteau nival est responsable de teneur en ions et en métaux dans les premières eaux de fusion, qui sont de 5 à 10 fois supérieures à celles qu'on observe dans la neige voisine. Les principaux ions ne sont pas éliminés du manteau nival à la même vitesse au cours de la fusion; ce processus est appelé "élution préférentielle". La chimie des eaux de surface au cours de la fusion est influencée principalement par la perte différentielle des polluants du manteau nival et par les voies empruntées par l'eau de fusion. Une diminution du pH, du PNA et des cations basiques survient, alors que les teneurs en Al et NO_3^- augmentent souvent, bien que le comportement de ce dernier ion soit variable d'un endroit à l'autre. L'effet des eaux de fusion est généralement limité à la couche voisine de la surface (épaisseur, 1-3 m) dans les lacs; toutefois, ce comportement est fortement influencé par plusieurs facteurs physiques. La fusion printanière peut avoir d'importantes conséquences pour plusieurs espèces de biotes aquatiques.

1. INTRODUCTION

In areas where accumulation of a winter-long snowpack is a normal phenomenon, rapid melting during spring almost always yields the major hydrological event of the year. In northern latitudes, these areas often spatially coincide with large regional or point-source emitters of acid precursors (SO_2 and NO_x) leading to the accumulation of a pollutant load within the snowpack. Sudden release of this load during melting may provide a chemical event that is also of great significance (Jeffries et al., 1979), particularly for aquatic biota that are either inherently intolerant to rapid changes in pH or are passing through sensitive life stages during the melt period (Gunn and Keller, 1984; Mierle et al. 1986). In some areas, drinking water quality may also be impaired (Liebfried et al., 1984). The short-term magnitude of the chemical event is accentuated by differential elution of ionic pollutants (relative to water) from the snowpack (Johannessen and Henriksen, 1978) although several other climatic, hydrologic, and terrain related factors can also be important in determining its ultimate ecological effect.

Late-winter sampling of the snowpack is a potential means of defining the regional pattern and magnitude of acidic deposition (e.g. Wright and Dovland, 1978). Sampling techniques are well established, having been developed earlier for the assessment of water equivalent and spring runoff forecasting (Colbeck, 1987a); furthermore, snowpack surveys are inexpensive compared to direct deposition monitoring. However, the utility of snow surveys is dependent on the assumption that the pollutant load is conservative, that is it exhibits little loss or internal alteration prior to sampling. Since many of the regions of concern commonly experience short term periods of melting in mid-winter, this assumption is often not valid, and therefore in practice, snowpack surveys have found only limited application.

A need to understand the factors governing the storage and release of pollutants from the snowpack and the resulting ecological effect is clearly reflected in the burgeoning scientific literature on the subject, for example see the review by Marmorek et al. (1984) and portions of Marmorek et al. (1986). Entire volumes are now being dedicated to defining the physics, chemistry and hydrology of snowpacks (Jones and Orville-Thomas, 1987). My objective here is to present a concise review of the most important findings associated with 3 aspects of snow and snowpack research, namely definition of (a) snowpack chemistry, (b) physical and chemical snowpack processes, and (c) snowmelt induced effects on surface water chemistry. Information on modelling of snowpack and snowmelt processes will be noted where appropriate, but in-depth discussion is beyond the scope of this chapter. Snowmelt effects on aquatic biota will also be mentioned briefly. While I have attempted to extensively cite important references to increase the utility of this chapter, it should not be construed as an exhaustive review; the 3 references immediately above already satisfy such a need.

2. SNOWPACK CHEMISTRY

The chemical content of the snowpack is influenced by many factors of which the most important are the magnitude of both wet and dry deposition, and the major impact of melting and/or rain-on-snow episodes. Other factors that may play locally important roles include: terrain physiography which may strongly influence snowfall amount, type of forest cover which influences the incorporation of natural organic debris during pack accumulation, wind induced snow redistribution, and existence of significant local sources.

Snowpack surveys, particularly those conducted on a regional scale, are designed to minimize the potential variability introduced by these factors. Hence sampling protocols usually demand that integrated snow core samples be taken well before there are above-freezing temperatures and at sites that both minimize the potential for pack alteration by organic debris or drifting (typically forest clearings) and are remote from urban or industrial influences (Barrie and Vet, 1984). Multiple coring at each site is often employed to overcome uncertainties arising from snowpack heterogeneity. Snow core surveys employing more intensive spatial and temporal sampling activity still use the essential elements of this protocol (e.g. English et al., 1987). In all cases, the very low concentrations of some chemical parameters (particularly the trace metals) demand that great care be taken during sample collection, processing, and analysis to ensure that no contamination occurs. This may involve approaching the sampling point by foot, using specially constructed and cleaned coring devices (generally made from plexiglas or other like substance), and wearing polyethylene gloves. Use of "clean laboratory" conditions for sample processing and analysis may also be appropriate. Even these precautions are inadequate for investigations at "background" locations (Legrand and Delmas, 1987).

Snowpack chemical sampling has been used to address a wide range of scientific questions. These range from definition of winter deposition in regional and sub-regional settings, to investigations of hydrochemical processes at specific locations. Regional surveys occur in locations having a widespread snowpack that is permanent for several months each year, usually in Scandinavia, Canada and portions of the northern USA. Snowpack studies in other locations are usually conducted only in alpine settings.

Compiling data from such diverse studies into a coherent and useable summary demanded that some consistent criteria for inclusion be established. I have not performed an exhaustive literature search and data compilation; rather I have attempted to summarize general regional situations. Therefore studies centered around major industrial point sources of pollutants have been excluded, e.g. data around smelters in Ontario, Canada (Jeffries and Snyder, 1981a), Manitoba, Canada (Phillips et al., 1986) or Poland (Zajac and Grodzinska, 1982). However, I have included data from the Montreal, Quebec area (Lewis et al., 1983; Landsberger et al., 1982) in order to demonstrate the influence of a major urban source. Only "pre-melt" snowpack chemistry was considered; however, when pre-melt data for both "new" and "old" snow were given in a reference (usually to demonstrate the influence of dry deposition), the "old" data were selected as they better represent the pollutant load that is released during subsequent snowmelt. Many publications present extensive data tabulations; in these situations, parameter means have been calculated for use in the following discussion despite the fact that substantial between-location variability often exists. Certain data presentations of snowpack composition (e.g. Wright and Dovland, 1978) were not amenable to statistical reduction for the purpose of this discussion due to their geographic (rather than tabular) style and therefore could not be included. In the specific case of Wright and Dovland (1978), a portion of their source data set (i.e. Gjessing et al., 1976) was used as a reasonable alternative. Finally, it should be noted that the data presented below are exclusively snow"pack" data. A large body of information exists for snow chemistry within the literature devoted to atmospheric deposition; consideration of it is beyond the scope of this chapter, however.

The summary of compiled data has been split into 3 categories, i.e. major ions, trace metals, and organic contaminants. Dewalle (1987) has also summarized snowpack data for ions and metals but has placed special emphasis on eastern North America.

2.1 Major Ions

Major ion concentrations in "regional" (one exception) snowpacks have been summarized in Table I. The most striking feature of these data is the large degree of variability that exists among regions and/or sites and the very large range of reported values - qualities in common with atmospheric deposition data. This large variability is present despite the fact that a significant portion of the overall raw data variability has been excluded by calculation of mean values as discussed above. For example, Barrie and Vet (1984) in a large regional survey that encompassed most of southern Ontario and Quebec in Canada, found that the mean within-sampling-site coefficient of variation (CV) for ionic parameters ranged from 12% (NO_3^-) to 41% (Cl^-). These CV values are remarkably similar to those reported by Jeffries and Snyder (1981a) in the Muskoka-Haliburton region of south-central Ontario (13% for NH_4^+ and NO_3^- to 45% for Cl^-) and inside the range reported for the Turkey Lakes Watershed in central Ontario (5% for NO_3^- to 151% for Na^+ ; Semkin and Jeffries, in press).

Marine influences on snowpack chemistry occur throughout Table I and these must be considered when attempting to relate observed composition to the long range atmospheric transport of pollutants. One location in Antarctica (d), the North West Territories location in northern Canada (g), and the Scottish (t,u), Norwegian (y,z), and the Japanese (cc) sites are all probably affected by the deposition of marine aerosols. This influence is most apparent in elevated concentrations of both Cl^- and Na^+ , but also must necessarily affect the snowpack concentrations of other major sea water constituents (i.e. Ca^{2+} , Mg^{2+} , and SO_4^{2-}). Estimation of ion concentrations in excess of the marine source is a routine calculation, e.g. Wright and Dovland (1978) corrected Norwegian survey data for this influence in assessing the SO_4^{2-} contribution from distant sources. It is probable that the Welsh site (s in Table I) also receives a significant marine input although the wide disparity in Na^+ and Cl^- concentrations (on an equivalent basis) suggests that an additional source of HCl is present, a hypothesis also advanced by Tranter et al. (1986) for the Scottish snowpack. The marine influence on ions other than Na^+ and Cl^- is often not obvious when viewing tabular data; however, at background locations such as Antarctica (d), the effect on Ca^{2+} , Mg^{2+} and SO_4^{2-} concentrations is apparent when the data are compared to other more inland background sampling locations.

Once the marine influences in Table I have been considered, the order of dominance of remaining ions can be determined for each sampling location. While this order is not absolutely consistent from location to location, dominant cations are usually H^+ , Ca^{2+} , and NH_4^+ while dominant anions are SO_4^{2-} and NO_3^- . As discussed by Tranter et al. (1986), H^+ , SO_4^{2-} , and NO_3^- concentrations are often closely related (often significantly correlated statistically) and indicative of acidic pollutant inputs. In a general way, snowpack concentrations of these 3 parameters in Table I reflect pollutant deposition levels. Hence background locations have exceedingly low values while the highest regional values occur in the north-eastern USA, southern Ontario and Quebec, Britain, and central Europe. Similarly, Wright and Dovland (1978) showed that the generally higher H^+ , NO_3^- , and sea-salt corrected SO_4^{2-} snowpack concentrations in the south of Norway compared to the north could be directly related to the long range input of acidic pollutants from the major European emission areas.

Specific exceptions to these generalizations can usually be traced to the influence of a local factor influencing the snowpack chemistry. For example, Jones (1984) sampled snowpacks located in a boreal forest setting in southern Quebec, and leaching of basic cations (particularly K^+) from the substantial quantity of organic debris that is normally

incorporated into the snow at this location was considered to substantially influence the pack chemistry. Statistical analysis of this data (i.e. factor analysis) clearly separated those ions primarily originating from long range transport, and those due to local deposition phenomena and organic debris (Jones, 1985). Similarly, Cadle et al. (1986) reported that snow collected under a red pine canopy had significantly lower H^+ and NO_3^- than snow in an open field; deciduous canopies exhibited negligible influences however. These exceptions cannot be taken lightly; the terrain most influenced by acidic deposition in North America and Europe is generally forested.

High strength but diffuse sources of pollutants such as the major urban center of Montreal, Quebec (Lewis et al., 1983; m in Table I) may profoundly affect snowpack chemistry, giving overall concentrations that are unlike any other surrounding regional data, and parameter-to-parameter relationships that reflect the local anthropogenic activity. High levels of Na^+ and Cl^- in the urban Montreal snowpack reveal not a marine influence but the extensive use of salt on urban streets for ice control; similarly, high SO_4^{2-} and NO_3^- show the influence of local space heating and automotive emissions. Elevated basic cation concentrations coupled with somewhat lower H^+ (than might be expected given the acid anion levels) probably reflect partial dissolution of the large load of natural and anthropogenic particulate material incorporated into the snowpack in an urban setting. Between snowfall-event dry deposition is likely a major contributor to the snowpack chemical load in this (or any) urban setting (Cadle and Dasch, 1987).

Sulphate to nitrate equivalent concentration ratios may provide information on the relative contribution of SO_2 and NO_x emission sources to the snowpack acidity. Using sea-salt corrected SO_4^{2-} concentrations, the median $SO_4^{2-}:NO_3^-$ ratios for regional data in Table I are 1.3, 0.8, and 1.4 for remote areas, North America, and Europe respectively. These ratios show that NO_x sources are a significant source of acidity in the snowpack in all locations, and in fact NO_3^- predominates over SO_4^{2-} in North America, an observation previously noted by several authors e.g. Johannes et al. (1980), Barrie and Vet (1984), Semkin and Jeffries (1986a). This interpretation contrasts with the fact that emission strengths for the latter are known to be greater, and the ratios, when calculated for annual deposition measurements, invariably show SO_4^{2-} as dominant. The occurrence of snowpack $SO_4^{2-}:NO_3^-$ ratios near or less than 1 arises due to the strong seasonal cycle in SO_4^{2-} deposition (high in summer, low in winter) and the comparative seasonal constancy of NO_3^- deposition (Semkin and Jeffries, 1986b; Summers and Barrie, 1986). Hypothesized reasons to explain the seasonal cycle in SO_4^{2-} range from decreased atmospheric SO_2 oxidation rates in winter (Summers and Barrie, 1986) to a reduced natural (biogenic) source of S during the winter (Nriagu et al. 1987).

2.2 Trace Metals

Snowpack concentrations for metals normally found in trace amounts in natural waters are presented in Table II. The quantity of published data is far more restricted for metals than it is for major ions, although there is a fairly substantial literature on metals in deposition (see reviews by Jeffries and Snyder, 1981b; Galloway et al., 1982). It should be noted that while Table II specifies "total" metal concentration, this is based on the methods descriptions, and methods of sample handling, processing and analysis vary greatly among the references. Certainly it is well established that sample contamination can occur easily for any low concentration parameters such as these. Measurement of metal levels typical of those reported for snowpacks in remote areas (Table II) require extraordinary and exacting methodologies (Legrand and Delmas, 1987).

Snowpack metal concentrations at regional sites in north-eastern North America and

Europe are generally 10 - 1000-fold greater than those reported for remote areas. This is true of all the metals in Table II, both those having a major lithologic source (i.e. Fe, Mn, Al) and those probably released by anthropogenic activities (see enrichment factor discussion in Jeffries and Snyder, 1981b, and Barrie and Vet, 1984). There is large variability in snowpack concentrations among the regional sites, but levels are generally similar to those found in deposition samples (Barrie and Vet, 1984). Intensive anthropogenic activities are reflected in the high metal concentrations found in the urban Montreal snowpack (Landsberger et al., 1982). The high levels of Fe, Mn, and Al support the earlier hypothesis of elevated "dust" fallout in the urban site.

2.3 Synthetic Organics

Information on snowpack concentrations of synthetic organic contaminants is extremely limited. The data in Table III for various pesticides and total PCB's are presented primarily for information. Data also exists for chlorophenols in Finnish snow (Paasivirta et al., 1985a, 1985b). Wide differences in sampling and analytical methodologies, and the resulting parameter precisions and detection limits preclude meaningful site-to-site comparisons. In general, pesticide and PCB concentrations are very low. It is perhaps significant that of available the data, many are for very remote locations, presumably having been collected to demonstrate the long range transport of these compounds. Clearly the paucity of data for synthetic organic contaminants and trace metals in snow points to a continuing research need.

3. SNOWPACK PROCESSES

3.1 Snowpack Stability

Chemical stability of the snowpack is an essential requirement (and assumption) of the regional surveys discussed above. In many locations near the geographic limits of uninterrupted winter snowpack development (areas that also often receive acidic deposition), the occurrence of a mid-winter thaw is the rule rather than the exception and this must be considered when assessing the accumulation of a pollutant load. Apart from the obvious degradation that accompanies above-freezing air temperatures, the experimental evidence on snowpack chemical stability suggests that many site-specific factors play a role. Furthermore, dry deposition to an existing snowpack can be important in increasing the chemical load present. It should be noted that the following discussion on snowpack "stability" refers only to preservation of its chemical character; the normal processes of physical metamorphism (within-pack flow of water vapour, snow crystal growth, and compaction) are not included here, even though Colbeck (1981) has shown that exsolving of chemical impurities to crystal surfaces occurs during re-crystallization. Snow metamorphism has been reviewed recently by Colbeck (1987b).

Dry deposition to a snowpack has been investigated through several field experimental studies. Cadle et al. (1984a) quantified dry inputs of ions by water extraction of the "dry-side" bucket of a wet-only Wong precipitation collector. This procedure, while providing apparently reasonable results, had a large uncertainty due to the problems known to exist from over-collection of particles in a bucket-like container (Ibrahim et al., 1983). Later studies (Cadle et al., 1985; Cadle et al., 1986) used specially prepared snow surfaces (snow contained in a 2 cm high bucket) to directly measure dry deposition from the difference in concentration between the pre- and post-exposure snow (exposure period was 3-4 days). An earlier study (Forland and Gjessing, 1975) also used short term changes (3 days) in the concentration of surface snow to infer dry deposition. Semkin and Jeffries (in press) estimated winter season dry inputs by comparing cumulative wet

deposition with the measured snowpack load, and later with cumulative snowmelt from a 1 m² snow lysimeter. The results of studies in rural locations by Cadle et al. (1986) and Semkin and Jeffries (in press) are shown in Table IV. The dry component of total chemical deposition to the snowpack is, in general, smaller than the wet component; however, there is substantial variability both between parameters and specific ionic species. The dry component is much more important near pollutant source areas (Dasch and Cadle, 1986). Figure 1 presents results reported by Semkin and Jeffries (in press); note that cumulative wet deposition of ions is consistently less than the observed snowpack ion content while cumulative bulk deposition is consistently greater than the snowpack content. Clearly, dry inputs significantly modify the overall pollutant load stored in the snowpack.

Several other studies have attempted to estimate dry deposition by determining deposition velocities (V_d) for the various chemical species to a snow surface (e.g. Ibrahim et al., 1983; Granat and Johansson, 1983; Bales et al., 1987). Multiplication of relatively easily measured air concentrations by V_d gives the dry input value. Discussion of the concept of a V_d , the methods used to determine it, and the factors influencing it, is beyond the scope of this review. However, it is important to note that V_d for some chemical species is strongly temperature dependent (particularly the gaseous S and N species) which helps to explain the wide range in values for H^+ , SO_4^{2-} , and NO_3^- in Table IV.

Various kinds of field evidence has been reported to demonstrate that a pre-melt snowpack can be chemically stable and therefore useful as a regional survey tool. Elgmork et al. (1973) carefully examined snow cores collected in southern Norway which contained visible grey layers that corresponded to high concentration snowfall events. They concluded that the snowpack was stable throughout the winter since a similar pattern of grey layers could be identified from core to core on a regional basis, and the ionic composition of corresponding layers was reasonably constant. Cadle et al. (1984a, 1984b) similarly examined the compositional constancy through time in 10 cm sub-sections of snow cores in N. Michigan. They also concluded that the snowpack is chemically stable prior to the first melt. Cadle et al. also inferred stability for both the pre-melt water and ion content through comparison of measured cumulative deposition with the standing load in the snowpack, an approach also used by Semkin and Jeffries (in press). Figure 1 demonstrates typical results; adding dry deposition to the cumulative wet deposition or correcting the cumulative bulk deposition for over-collection of the dry component (Ibrahim et al., 1983) yields nearly coincidental pre-melt curves for ions, e.g. similar to water. Finally, analysis of the isotopic composition of the snowpack profile through time in Switzerland (Stichler et al., 1981) demonstrated chemical stability throughout the winter in an alpine location, an observation later corroborated for various ionic species but not for H_2O_2 (Sigg et al., 1987). The loss of H_2O_2 depended mainly on the availability of sunlight suggesting that any photodegradable compound (e.g. various organic contaminants) similarly may not be stable in snow.

There is also literature demonstrating pre-melt instability of snowpacks. Schemenauer et al. (1985) presents snowpack temperature profiles for a central Ontario location. In late January, temperature increased uniformly from -7°C at the surface to near 0°C at the ground-snow interface. This situation is typical of this area; heavy snow in early winter usually insulates the ground from freezing. In such a situation, a convective heat flux from the underlying soils can cause substantial water equivalent loss from the snowpack (Motoyama et al., 1986; English et al., 1987). Loss of ions at the same time may explain the observation of pre-melt snowpack alteration reported by Jeffries and Snyder (1981a) and Babiakova and Bodis (1986). Jones and Bisson (1984) also reported loss of ions from

a southern Quebec snowpack prior to the onset of melting. Like the heat diffusion and water vapour movement evidently occurring within the snowpack, gases generated from soil microbiological processes (particularly those in unfrozen soils) are no doubt influencing the pack; however, I can find no literature describing studies along this line. Jones (1987) does show that NO_3^- and NH_4^+ loss from the snow in a boreal forest site in southern Quebec is associated with microbiological activity on the organic debris from the forest canopy; further discussion can also be found in Jones (1985) and Jones and Sochanska (1985). Finally, Page (1987) reported cases of ion movement in alpine snowpacks in France although it is not clear how much of this can be attributed to melt episodes; perhaps of greater interest is his speculation of the upward movement of ions from the soil into the snow.

The complexities of assessing snowpack stability is apparent when it is noted that the observations of English et al. (1987) were made in the Turkey Lakes Watershed, the same location as the observations of Semkin and Jeffries (in press) though specific sampling sites were not identical. The conflicting interpretations of snowpack stability from these co-located studies points to poor understanding of the multiple confounding factors controlling these observations and the spatial heterogeneity that probably exists. Clearly further research is warranted in this regard.

3.2 Snowpack Hydrology

Understanding melt generating mechanisms and within snowpack water flowpaths is essential for predicting the influence of snowmelt on associated terrestrial and aquatic ecosystems. Several evaluations of snow surface energy exchange (e.g. Male and Granger, 1981) often in the context of model development (Goodison et al., 1986; Price, 1987; Stein et al., 1987) show that the net radiative energy flux is most responsible for snowmelt production. The ideal case of simple gravity flow of meltwater through the snowpack has been described (Colbeck and Anderson, 1982; Jordan, 1983a) and several predictive models developed (e.g. Jordan, 1983b). Under such conditions, meltwater movement is rather slow (Jordan 1983a reported average meltwater wave front velocity of 0.22 m h^{-1}) and chemical exchange and homogenization among the snowpack stata will occur (e.g. the isotopic evidence presented by Stichler et al., 1981).

Snowpack delivery of meltwater is complicated by several other factors however. Diurnal freeze-thaw cycles can delay the appearance of meltwater at the base of the pack and models have been modified to account for this (Bengtsson 1982a, 1982b). Ice or high density snow layers contained within the pack can strongly influence the spatial delivery of meltwater. For example, English et al. (1986, 1987) have shown that substantial lateral diversion of meltwater occurred in a hillside snowpack in central Ontario; the net result being that the lower portions of the hillslope receive a greater water and chemical loading than the upper portion. Marsh and Woo (1985) similarly observed the strong influence of ice layers on ground level meltwater delivery in the Arctic, and have developed a descriptive multi-flowpath model. Jones (1985) has shown that various mesostructural characteristics of the snowpack (including ice layers) can lead to very heterogeneous meltwater flow patterns. In particular, pipeflow can develop around organic debris (twigs, shrub and tree stems, etc) that is capable of rapidly delivering a large proportion of the meltwater to spatially limited locations at the ground-snow interface; also, vertical pipeflow develops within the drip zone of the forest canopy. Furthermore, as the pack becomes saturated, the snow mesostructure can change very rapidly. These observations point out the difficulty that exists in interpreting data on spring melt snowpack decay in the non-ideal but normal field situation.

Rainfall can also strongly influence water delivery from a melting snowpack by dramatically increasing its free water content and thereby altering the normal flow patterns (not to mention any rain induced mesostructural changes as above). Semkin and Jeffries (1986a) reported that rainfall caused an almost immediate response in water delivery from a snow lysimeter, and Brown et al. (1985) used isotopic analyses to show that almost all the water leaving a snowpack after rain events was, in fact, rain water.

3.3 Chemical Loss from the Snowpack

Original concern regarding the snowpack release of pollutants arose from the groundbreaking paper of Johannessen and Henriksen (1978). In it they described the fractionation of pollutants in the snow and occurrence of high concentrations of both ionic and metal species within the early melt fractions. The physical processes yielding these observations were described by Colbeck (1981). During normal snowpack metamorphosis involving grain coalescence and recrystallization, impurities within the snow tend to segregate at the grain surfaces because they are not easily incorporated into the crystalline lattice. Later when liquid water begins to pass through the snowpack, these segregated impurities are readily dissolved and appear in high concentrations in the first melt fractions. The entire process can be repeated with subsequent freeze-thaw events, each displaying high pollutant levels in the early melt; however, the absolute magnitude of the concentration maxima must necessarily decline with each event. Colbeck also notes that the rate at which the impurities are removed depends on the atmospheric conditions under which the snow was deposited, the degree and type of snow metamorphism, and the sequence and/or intensity of the liquid water supply, be it melting or rainfall. There have been many publications verifying the occurrence of this differential fractionation and release of pollutants (e.g. Johannes et al., 1980; Cadle et al., 1984a; Semkin and Jeffries, 1986a, in press; Babiakova and Bodis, 1986; Tranter et al., 1986; Brimblecombe et al., 1987; Davies et al., 1987; there are many others). Models have also been successfully developed to describe this phenomenon (Leung and Carmichael, 1984; Hibberd, 1984).

Typical examples of ion fractionation and release for H^+ , SO_4^{2-} and NO_3^- are illustrated in Figures 2 and 3. The data are for the 84/85 and 85/86 snow seasons respectively in the Turkey Lake Watershed (Semkin and Jeffries, 1986a; Semkin and Jeffries, in press) and were obtained using a field-based snow lysimeter; hence the snowmelt values reflect the sum of both meltwater and rainfall. The 3 ions presented in Figures 2 and 3 clearly exhibit high early meltwater concentrations relative to the parent snowpack; concentration factors are approximately 10-fold for H^+ and SO_4^{2-} and approximately 7-fold for NO_3^- . These concentration factors are somewhat higher than both those reported by Cadle et al. (1987) for a northern Michigan snowpack (i.e. approximately 5-fold ion enrichment) and the 6.5-fold value reported by Johannessen and Henriksen (1978). Comparison of the pattern of meltwater concentration between years also illustrates the influence of rainfall. The 1985 snowmelt season at the Turkey Lakes experienced several substantial rainfall events, whereas the comparatively short, warm melt period in 1986 was relatively rain free. Hence, in 1985, multiple concentration peaks in the lysimeter "meltwater" output can be directly related to the input of rain, and the virtually instantaneous concentration response at the time of the event corroborates the observations of Brown et al. (1985) that rain often passes directly through the snowpack without substantial interaction. Considering the entire 1985 melt period, rain accounted for 18% of the water volume and 51%, 49%, and 37% of the H^+ , SO_4^{2-} , and NO_3^- snowpack outputs respectively. Meltwater concentration versus time plots for the low rain 1986 melt season (Figure 3) look remarkably like those presented by Johannessen and Henriksen (1978); rain contributed only 14% of the water and 12%, 22%,

and 16% of the H^+ , SO_4^{2-} , and NO_3^- output. It should be noted that Johannessen and Henriksen (1978) only presented those experiments in which rain was not present, rightly expecting that it would confound the simple phenomenon they were trying to illustrate.

The differential process of impurity concentration and release has been termed "preferential elution". There is a sequence of ion elution as illustrated in Figure 4, i.e. the sequence from the Turkey Lakes Watershed snowpack is $SO_4^{2-} > NO_3^- > H^+ > Cl^-$. The percent of total ion output as a function of water output can also be read directly from Figure 4. The elution sequence is not unique; Tranter et al. (1986) reported similar results for the Scottish snowpack, and in fact, the comparatively high sea salt component in this location tends to accentuate the development of the late eluting NaCl fraction. Data presented by Babiakova and Bodis (1986) in Czechoslovakia are remarkably similar as well. The underlying reason for the elution sequence was noted by Colbeck (1981) and has been extensively studied under both field and laboratory conditions; Davies et al. (1987) present a substantial review discussion of this subject. Briefly, those ions which are more soluble in ice (i.e. incorporate more easily into the ice crystal lattice) are the least mobile upon melting. In contrast, the earliest eluting ions are those which are most strongly partitioned to the ice crystal surface during the fractionation process accompanying snow metamorphosis.

So-called "acid shock potential" models have been developed that use a simple empirical expression of the differential release of snowpack acidity (Wilson and Barrie, 1981; Agnew et al., 1982; Goodison et al., 1986). These models generally have the intent of predicting acid shock potential using a climatic probability scheme and employ an exponential factor to simulate the differential elution of acidity. The numeric value of the factor is usually based on the results presented by Johannessen and Henriksen (1978).

4. SNOWMELT EFFECTS

Preferential elution from the snowpack lies at the heart of concern for springmelt pH depression in surface waters and the potential impact it may have on aquatic organisms. There are only a limited number of biogeochemical mechanisms by which an ecosystem can successfully counteract the rapid input of highly acidic meltwaters.

4.1 Surface Water Chemistry

The differential release of acids during snow melt occurs at a time when the runoff waters have a reduced ability to neutralize them. Even in the absence of an elevated pollutant load in the snowpack, spring melt is a major hydrochemical event resulting in rapid changes in the concentration of many chemical species due to both dilution and the exploitation of flow pathways associated with high water fluxes. Pathways play an important role in controlling the eventual runoff chemistry, and it is useful to provide a brief summary of the hydrological characteristics and soil properties that bear on the development of water quality.

During low flow (i.e. the baseflow condition), water reaches a stream channel by flow through deeper soil horizons and/or porous bedrock. The contact time between water and geological material is therefore long and the water chemistry reflects the chemical properties of the matrix material; it almost invariably has a higher pH than the input precipitation. During spring snowmelt, "quickflow" hydrological mechanisms become important. Quickflow refers to water that is rapidly transmitted through the soils yielding a sharp rise in the stream hydrograph. In order for quickflow to occur, the soil

matrix delivering the water must have both a high hydraulic conductivity and a hydraulic gradient steep enough to foster a high flow rate. Snowmelt induced quickflow can be generated by several mechanisms; however, the most important are "return flow" and "subsurface stormflow". The former is caused by a rise in a near surface water table in response to increased hydrostatic pressure exerted by meltwater penetrating an adjacent unsaturated zone. The latter refers to rapid lateral movement of water through a subsurface (though usually near surface) zone that has a high saturated hydraulic conductivity. This zone, which normally is unsaturated during periods of low input but still contains significant amounts of pore water, may be quickly saturated during high input due to restriction of downward flow by underlying materials of low permeability (bedrock or lower soil layers). Peters and Murdoch (1985) and English et al. (1987) both present convincing field evidence of the occurrence of subsurface stormflow through relatively acidic near-surface soil layers. Pore waters previously resident in such layers are pushed out in a piston effect. Both of these quickflow mechanisms cause the appearance of "old" or "pre-event" subsurface water in the stream during snowmelt as has been reported several times by investigators using isotopic hydrograph separation techniques (e.g. Rohde, 1981; Bottomley et al., 1986; Lawrence, 1987). Subsurface pipeflow through tree root channels or the like can also be important during periods of high water flux and serve to confuse the interpretation of the snowmelt hydrology. More complete discussions of streamflow generation during snowmelt can be found elsewhere (Dunne, 1978; Price and Hendrie, 1983; Hendershot et al., 1984; McDonnell and Taylor, 1987).

Chemical alteration of the water leaving the snowpack within the terrestrial basin to yield what is eventually observed in the streams is a function of (among others) the permeability of the underlying materials, the amount of underlying soil material (i.e. controlling the overall potential for soil-water interaction), the geochemistry of the soils, the water input rate, and the quickflow flow path (e.g. water flowing laterally through organic or "A" soil horizons will achieve a different final composition from those flowing through "B" horizons). In the event that the substrate immediately under the snowpack is impermeable (or at least incapable of passing the water at the rate supplied), then overland flow will occur and the water reaching the stream channel will exhibit a composition that reflects the ion fractionation mechanisms discussed above. In fact, such cases have been observed (e.g. Skartveit and Gjessing, 1979; Johannessen et al., 1980) in which catchment outflow H^+ and SO_4^{2-} in Norway exhibits peaks in concentration that precede hydrographic peaks. This effect is due to the extremely thin to absent soils that overlay massive silicate bedrock in much of Norway. Similar results have also been reported for upland terrain in Scotland (Morris and Thomas, 1985).

Soil permeability at the snow interface can also be influenced by freezing, a condition that takes 1 of 2 forms, i.e. "honeycomb" or "concrete" (Post and Dreibelbis, 1942). The former is the normal condition in most temperate, forested terrain (Price and Hendrie, 1983) and leads to no significant decrease in the soil infiltration capacity; on the other hand, the latter normally occurs in sub-arctic and arctic climates (English, 1984) and often produces dramatic overland flow during spring melt. These situations are of lesser concern as far as this chapter is concerned, however, since the arctic snowpack generally contains only a small load of acidic pollutants. There are a few reported observations of the occasional development of concrete frost in temperate areas leading to the direct discharge of snowmelt waters into streams via overland flow (Price and Hendrie, 1983; Pierson and Taylor, 1985). These reports are the exceptions rather than the rule, requiring the presence of very wet soils during freezing periods; such conditions may occur with some frequency in boggy terrain, or may develop in response to heavy winter

rain followed by severe cold. Rainfall reduces the insulating properties of a snowpack (Schemenauer et al., 1985) thereby allowing development of concrete ice layers at the snowpack base (Price and Hendrie, 1983).

Overall, most of the temperate forested terrain in Europe and North America that is threatened by potential snowmelt effects experiences snowpack and soil conditions that permit easy infiltration of meltwaters. The subsurface stormflow occurring to produce the rapid peaks in stream discharge that are usually observed at the same time still permits significant chemical interaction of the meltwaters with the soil matrix within the water pathway. In particular, ion exchange reactions proceed very rapidly relative to the rate of water movement for even the most extreme cases of subsurface flow. Hence it is not surprising that under such conditions, the degree and timing of the pH depression is related more to the water flux than to the onset of melting, i.e. H^+ peak concentrations often occur well after the initiation of melting and are often coincident with hydrographic peaks (Jeffries et al., 1979; 1 of 3 cases presented by Johannessen et al., 1980; Siegel, 1981; Jeffries and Semkin, 1983; Bjarnborg, 1983; Cadle et al., 1984; Jacks et al., 1986).

Meltwater undergoes immediate and large changes in composition in those areas where soil infiltration predominates during snowmelt (Barry and Price, 1987). The chemistry of the receiving stream waters exhibits large reductions in alkalinity (or acid neutralizing capacity = ANC) as flow increases. While short term increases in base cation (Ca^{2+} , Mg^{2+} , Na^+ , K^+) concentrations may occur in streams at the initiation of melt as pre-event soil pore waters are pushed out (Johannessen et al., 1980), a substantial portion of the ANC reduction is simply due to dilution as reflected in the substantial reductions in base cations that subsequently occur (Galloway et al., 1980; Semkin and Jeffries, in press); however, some of it is usually attributed to the presence of strong acids as reflected in NO_3^- and SO_4^{2-} concentrations and an excessive depression in pH. Decreases in pH are often on the order of 1 pH unit or less (Jeffries et al., 1979; Galloway et al., 1980; Siegel, 1981; Keller, 1983; Bjarnborg, 1983; Henriksen et al., 1984) although there are examples of much more dramatic changes, e.g. Jacks et al. (1986). The stream pH values rarely ever approach the early snowmelt fraction pH. The relative influence of NO_3^- or SO_4^{2-} in producing the pH depression is often evaluated by consideration of the temporal variability of these ions. For example Galloway et al. (1980) noted that reduced pH corresponded with increased NO_3^- in the Adirondaks while SO_4^{2-} remained relatively constant, albeit at much higher overall concentrations than NO_3^- . The implication is that NO_3^- is strongly influencing the occurrence of the pH depression at this location, but is doing so under the influence of a greater and relatively constant acidification by SO_4^{2-} . The relative constancy of SO_4^{2-} reflects the rapid exchange processes that occur in the soil environment (Dahl et al., 1979); SO_4^{2-} adsorbed to soil particles during earlier portions of the year must be easily released as the dilute meltwaters pass through. This is likely the only explanation that allows both the SO_4^{2-} concentration "buffering" in the streams and still permits balanced annual input-output budgets for many catchments in glacial terrains (Jeffries et al., in press). The behavior of NO_3^- concentrations during spring melt is much more variable from location to location. In contrast to Galloway et al. (1980), Cadle et al. (1987) observed only a minor increase in NO_3^- at the beginning of the melt which rapidly decreased to near zero as the melt progressed; the differences are most likely related to the variable nutritional requirements for N exerted by the forests in question. If NO_3^- input is retained within the catchment, then it acts as a net source of ANC rather than a sink, and in fact, most basins do retain a significant portion of their total NO_3^- on an annual basis (Hemond and Eshleman, 1984). The high input rate and relatively low

biological production present during the melt period probably accounts for those observed cases of increased stream concentrations; however, as already noted, this is not a universal phenomenon.

Streams often exhibit elevated concentrations of Al during snowmelt (see review by Jeffries and Hendershot, in press). This phenomenon reflects both the routing of subsurface stormflow through near surface soil horizons with attendant ion exchange (Hendershot et al., 1985) and desorption of Al from the stream bottom substrate (Norton et al., 1987). In chronically acidified but not yet acidic catchments, total Al concentrations often increase from 10's to 100's of $\mu\text{g L}^{-1}$ although there are several reports of concentrations exceeding 1 mg L^{-1} . Such levels may be dangerous to aquatic biota although the concentration of the inorganic monomeric fraction is a better indicator in this regard. Henriksen et. (1984) reported fish mortality during spring runoff in the Vikedal R. of Norway in which the inorganic monomeric Al species increased to only $50 \mu\text{g L}^{-1}$.

Several models of varying complexity have been developed to predict short-term changes in catchment hydrology and runoff chemistry (e.g. Christophersen et al., 1983; Chen et al., 1983; Christophersen et al., 1984; Bergstrom et al, 1985; Lam et al., 1986; there are many other references). The effectiveness of these models is often tested by their ability to simulate the rapid changes in flow and chemistry that accompany spring melt. Compartmentalizing the subsurface flow hydrology (number of compartments varies from model to model) permits simulation of stream flow generation. Many of them employ the "mobile anion" concept as the main mechanism for transporting cations through soils into streams; adsorption-desorption, ion exchange, and weathering reactions are usually included. The models provide realistic simulations of snowmelt conditions in catchment outflows, and have been a most useful tool for integrating the many divergent and often competing processes operating at this time of year.

As with streamflow generation, the chemical effect of snowmelt on lakes is primarily controlled by the physical processes of meltwater dispersion under ice. Studies investigating these processes show that drainage basin characteristics (size and location of stream inflows), lake morphology, and residence time as well as climate (melt rates, rainfall, etc) are important in determining meltwater dispersion (Bergman and Welch, 1985; Bengtsson, 1986). The lower density of cold runoff waters (relative to the under-ice waters) means that the portion of the lake affected by snowmelt is restricted to a relatively narrow layer immediately under the ice; the incoming waters spread out across the lake and much of the snowpack output may in fact exit the lake via the outflow without significant interaction. Thickness of the under-ice layer is very often on the order of 1 m (Gunn and Keller, 1985; Bergman and Welch, 1985) although other studies report some interaction to a depth of approximately 3 m (Jeffries and Semkin, 1983; Hasselrot et al., 1987). Hasselrot et al. (1987) do report an interesting case of reverse layering (L. Bredvatten, Sweden) although it was spatially limited and due to the coincidence of specific physical factors.

Chemical composition of the spring melt runoff layer in the lake is very similar to that observed in the streams. Compared to pre-melt lake water, reduced pH, ANC, and base cations are observed (Jeffries and Semkin, 1983; Charette et al., 1984) while elevated NO_3^- and Al may also occur (Hasselrot et al., 1987; Jeffries and Hendershot, in press). Spatial heterogeneity is often observed in the layer due to the location of inflows and the influence of groundwater seepage. Another feature of spring inputs to lakes is the material trapped in the ice and snow laying directly on the lakes' surfaces. Ice decay

and eventual incorporation of these chemicals occurs at such a time (i.e. late spring) so that they are not immediately flushed from the lake. This input has been shown as important for the nutrient budgets (particularly P) of some lakes (Adams et al., 1979; Premo et al., 1985) and explains the swift rise in primary production that often occurs in lakes soon after ice break-up (Rask et al., 1985).

4.2 Aquatic Biology

A substantial literature exists describing the impact of snowmelt induced changes in surface water chemistry on aquatic biota; however, it is outside the scope of this chapter to review this material. Therefore, the following short description with citation of important references is primarily included for information and to identify the other major and active area of snowmelt research.

Certainly, one of the principal driving forces behind early acidic deposition investigations was to determine the cause of fish population loss and observed fish kills in lakes and rivers. Much of the early work was centered in Scandinavia. In particular, dramatic fish kills were often associated with spring runoff (Leivestad and Muniz, 1976; Henriksen et al., 1984), and pH and Al levels in the water appear to be the most critical factors (Baker and Schofield, 1982). Damage to fish populations by acidic deposition has been reviewed by Rosseland et al. (1986) for Scandinavia and by Haines and Baker (1986) for the northeastern US. In Canada, Harvey and Whelpdale (1986) demonstrated that snowmelt runoff in south-central Ontario causes fish mortality, and Gunn and Keller (1984) have shown that lake trout sac fry may be affected by spring melt since spawning beds are characteristically located in shallows within the acidified lake layer discussed above. Finally, a whole spectrum of non-fish aquatic biota are also susceptible to the influence of acidic deposition (reviewed by Mierle et al., 1986). Marmorek et al. (1984, 1986) provide comprehensive reviews of the effects of snowmelt and other episodic chemical events on aquatic biota.

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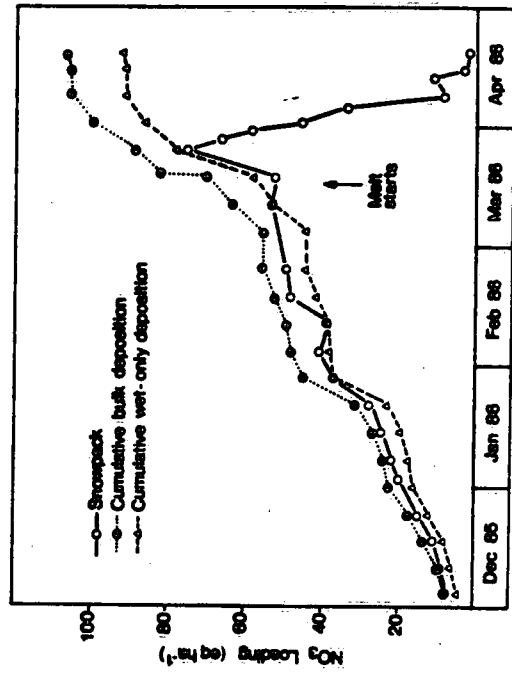
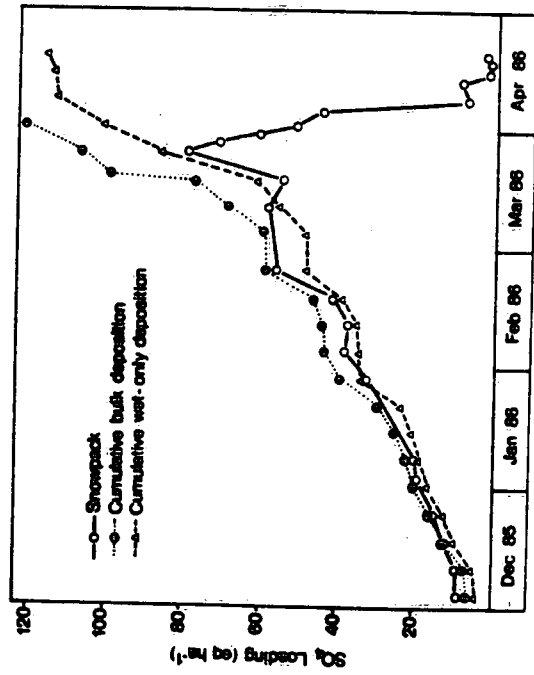
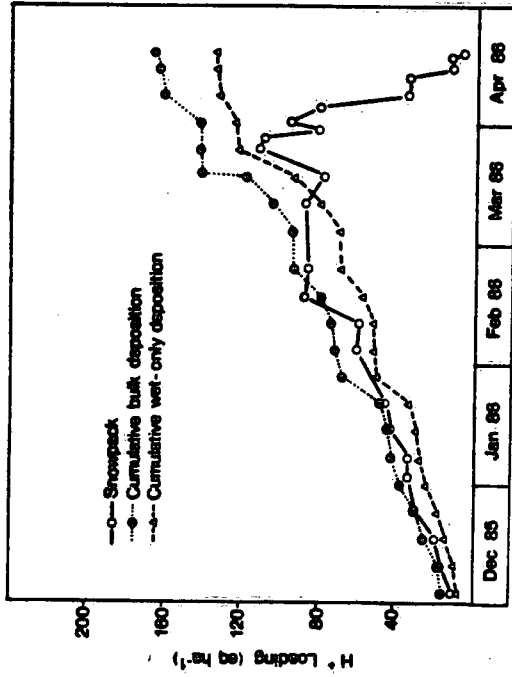
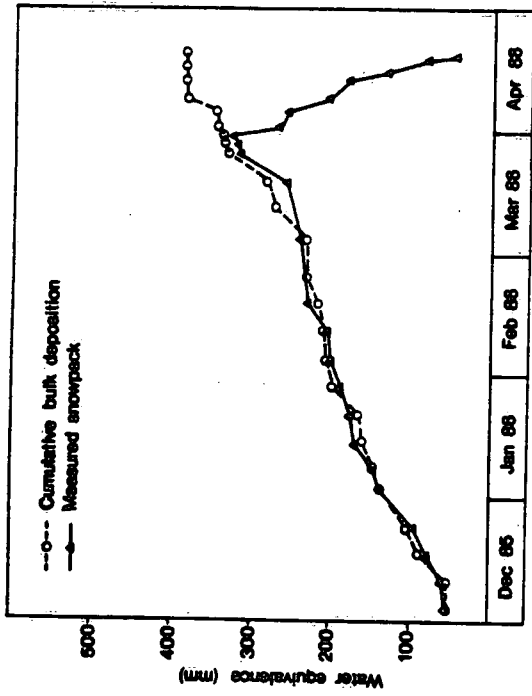
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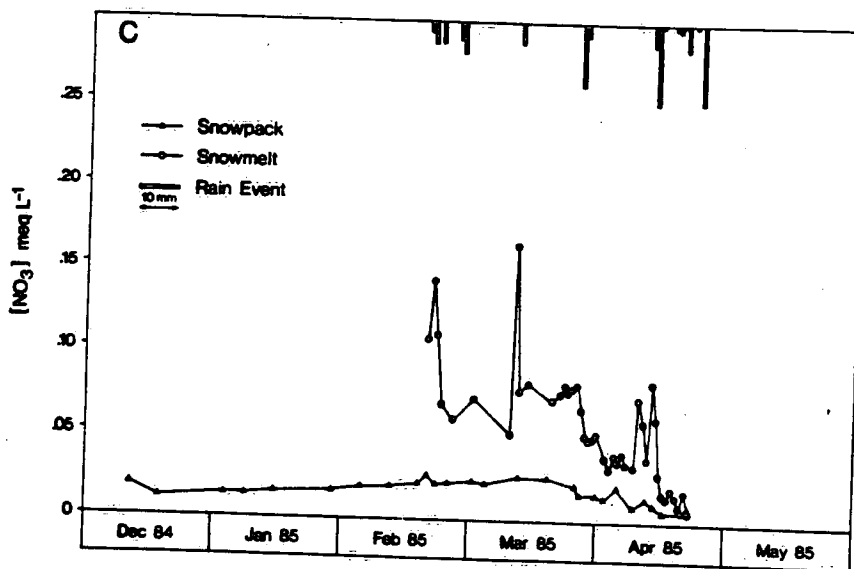
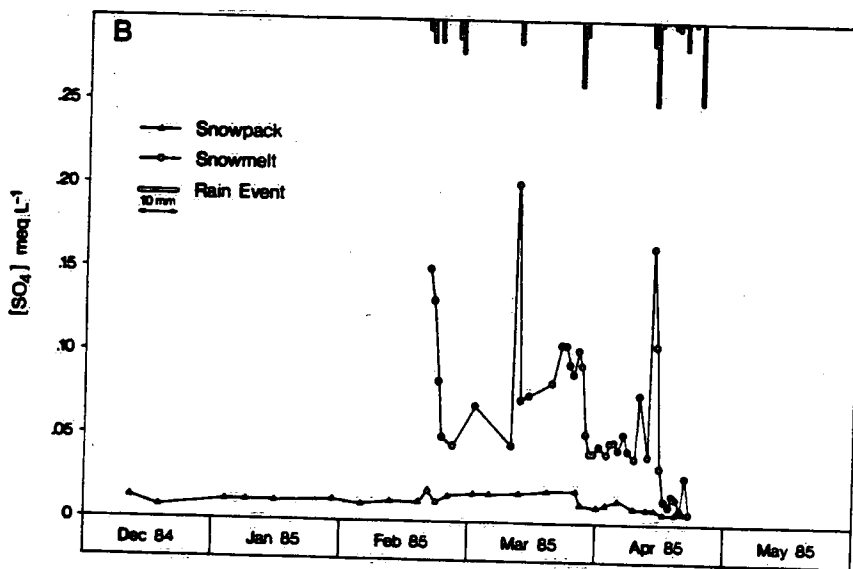
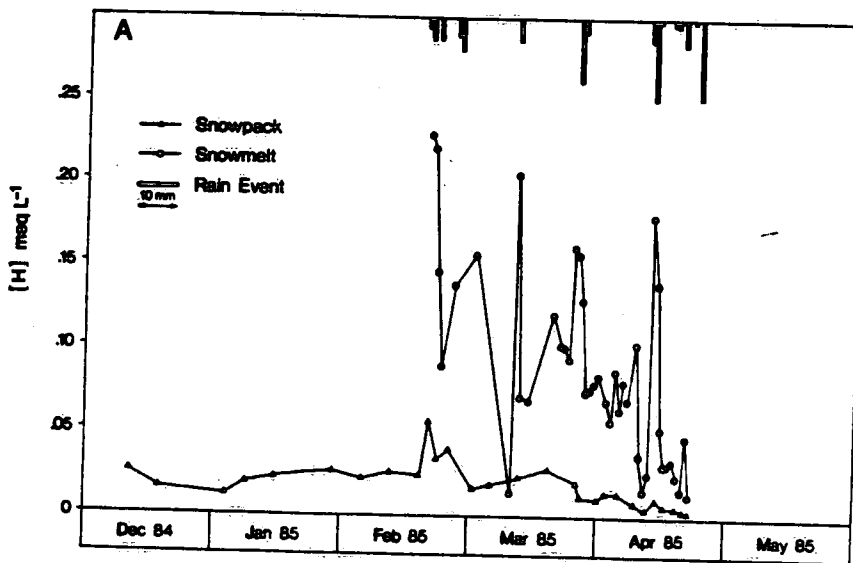
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LIST OF FIGURES

- Figure 1: Comparison of mass loadings of water (mm), and H^+ , SO_4^{2-} , and NO_3^- ($eq\ ha^{-1}$) in the snowpack with cumulative atmospheric deposition (both bulk and wet-only) for the 1985/86 winter season in the Turkey Lakes Watershed (after Semkin and Jeffries, in press).
- Figure 2: Concentration of H^+ , SO_4^{2-} , and NO_3^- ($meq\ L^{-1}$) in snowmelt (snow lysimeter output) and the parent snowpack through the 1984/85 winter season in the Turkey Lakes Watershed. The occurrence of rain events is also indicated. Note the lengthy melt period interrupted by significant re-freezing, and the marked influence of the rainfall events. Initial snowmelt concentrations are 5- to 10-fold greater than the levels in the parent snowpack (after Semkin and Jeffries, 1986a).
- Figure 3: Concentration of H^+ , SO_4^{2-} , and NO_3^- ($meq\ L^{-1}$) in snowmelt (snow lysimeter output) and the parent snowpack through the 1985/86 winter season in the Turkey Lakes Watershed. The occurrence of rain events is also indicated. Note the short melt period compared to 1984/85 and the minimal influence of rainfall (after Semkin and Jeffries, in press).
- Figure 4: Percent loss of H^+ , SO_4^{2-} , NO_3^- , and Cl^- as a function of percent water loss from the snowpack in the Turkey Lakes Watershed during the 1986 snowmelt. Preferential elution of the ions from the snowpack ($SO_4^{2-} > NO_3^- > H^+ > Cl^-$) accounts for the lack of coincidence of the lines. Approximately 50% of the major ion loading was lost from the snowpack with the first 20% of melt in 1986 (after Semkin and Jeffries, in press).





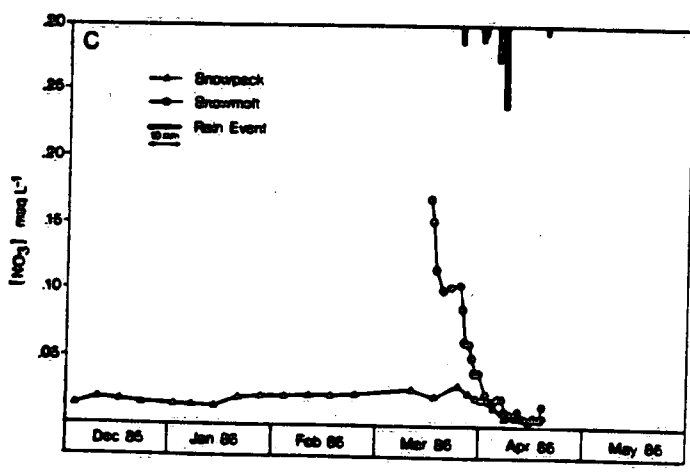
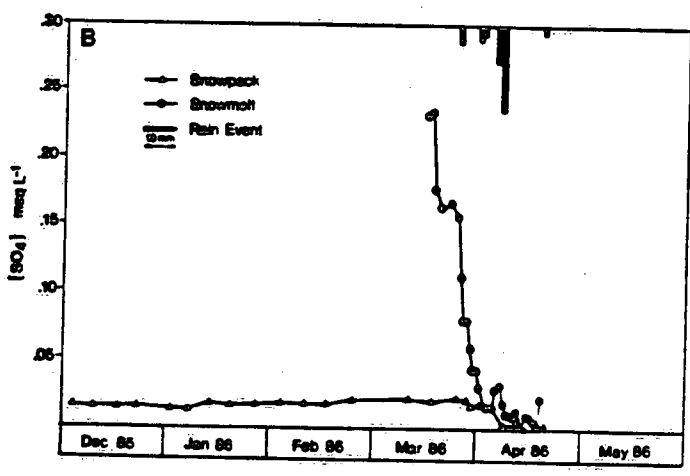
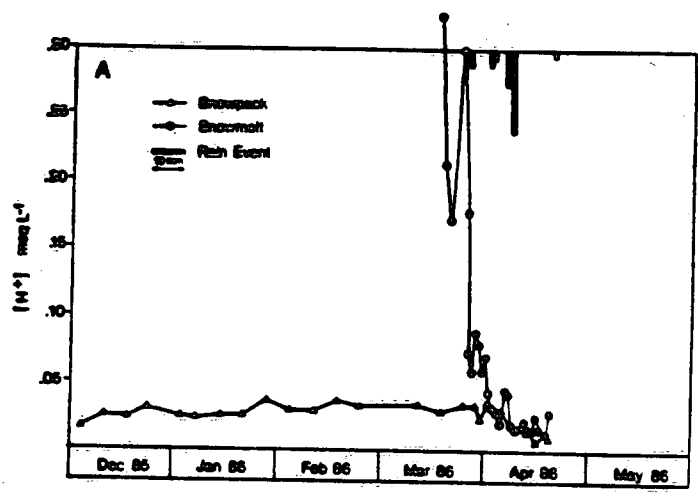


Fig 4

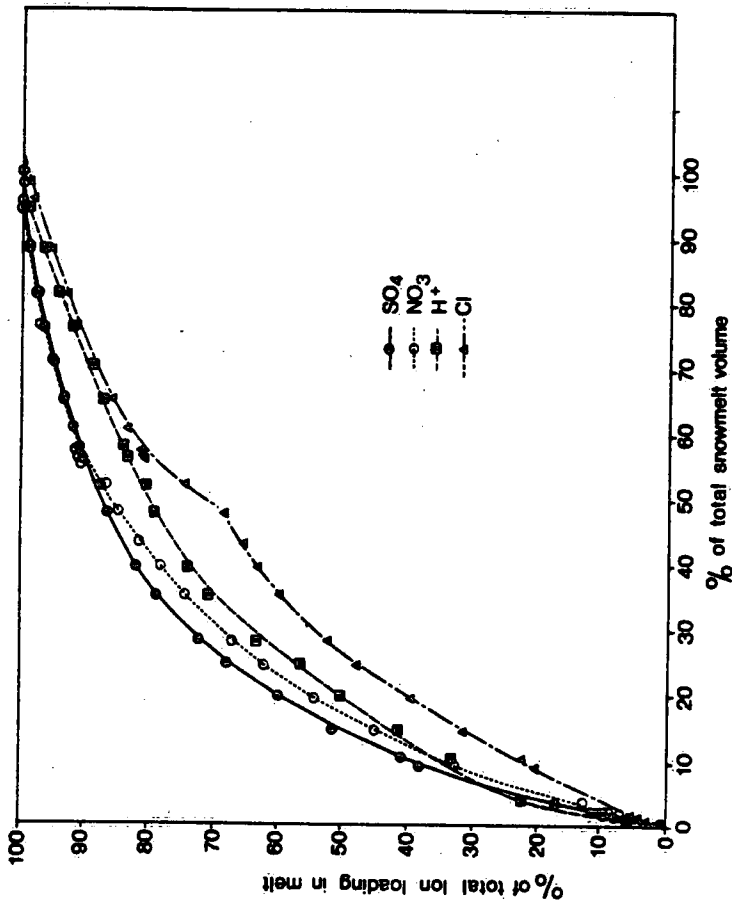


Table I: Major ion concentrations ($\mu\text{eq L}^{-1}$) in pre-melt snowpacks from remote, North American, European and Japanese locations. Data are mean or mid-range of values selected from each reference. "Less than" data in the references were taken at the nominal values when determining means; such means are indicated by "<". Consult text for other data selection criteria.

Location	H ⁺	Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	NH ₄ ⁺	SO ₄ ²⁻	Cl ⁻	NO ₃ ⁻
Remote Areas									
Ellesmere Island ^a	5.6			1.1					
Greenland ^b							1.9		1.4
Greenland ^c		<0.4	0.1	0.3	0.0		3.6		
Antarctica ^d		1.5	7.7			0.9	3.5	26.2	0.6
Antarctica ^e	3.4		0.2	0.6	0.0	0.2	1.5	1.3	1.4
North America									
British Columbia ^f	7.2	1.5				1.9			4.1
NW Territories ^g	22.4	6.8	3.1	11.7	<0.5		<13.8	12.4	
NW Ontario ^h	13.1	18.	5.7	10.0	2.4	2.8			14.7
Central Ontario ⁱ	24.6	4.4	1.6	4.7	0.6	8.1	12.3	4.5	21.8
S-Central Ontario ^j	45.0	14.				13.5	19.0	6.8	36.
South Quebec ^k	33.2	3.9	1.5	2.7	5.0	5.8	24.2	11.7	26.7
Eastern Canada ^l	40.1	3.6	<0.9	<2.1	0.6	8.8	18.1	<3.5	33.1
Montreal (urban) ^m	33.1	66.3	11.6	97.8	16.7	12.5	74.0	106.	50.2
Utah ⁿ	0.7						22.8		7.1
Minnesota ^o	17.7	8.0	1.5	6.1	3.1	7.9	14.4	6.8	16.4
N Michigan ^p	27.7	7.6	4.3	13.1	1.0	6.1	17.0	10.3	21.1
Pennsylvania ^q	55.9	62.4	9.9	17.4		19.1	36.8	22.3	64.6
New York ^r	54.						50.1	<0.3	45.8
Europe									
Wales ^s	129.	14.	11.	30.	5.		78.	69.	64.
Scotland ^t	31.6	3.5	13.	52.3	1.3		50.	28.8	5.5
Scotland ^u	137.		18.	76.4			55.3	111.	40.5
France ^v		5.8	4.0	2.4	3.1				
FRG ^w	52.							33.9	58.1
Czechoslovakia ^x	26.	54.	13.	7.2	7.5	24.	115.	36.	40.8
S Norway ^y	48.0	27.	8.8	50.6	13.	36.	65.	62.	39.
Norway (S of 65°) ^z	15.1	1.0	6.6	28.3	1.8	4.7	14.	27.	9.4
Norway (N of 65°) ^z	11.3	5.6	23.	108.	3.4	2.4	25.	110.	4.7
N Sweden ^{aa}	4.	13.		50.	<10.	4.5	17.	27.	4.
N Sweden ^{bb}	30.0	2.	3.	12.		0.8	23.	12.	17.
Japan ^{cc}		30.	39.	137.	5.8				

a Koerner and Fisher (1982), b Neftel et al. (1985), c Davidson et al. (1981), d Gjessing (1984), e Legrand and Delmas (1984), f McBean and Nikleva (1986), g Welch and Legault (1986), h Barica and Armstrong (1971), i Semkin and Jeffries (1986), j Muskoka-Haliburton only, Jeffries and Snyder (1981), k Jones (1984), l Barrie and Vet (1984), m Lewis et al. (1983), n Messer (1983), o Munger (1982), p Cadle et al. (1984), q DeWalle et al. (1983), r Galvin and Cline (1978), s Reynolds (1983), t Thomas and Morris (1985), u Tranter et al. (1986), v Batifol and Boutron (1984), w Schrimppff (1980), x Babiakova and Bodis (1986), y Johannessen and Henriksen (1978), z Gjessing et al. (1976), see also Wright and Dovland (1978), aa Bjarnborg (1983), bb Ross and Granat (1986), cc Suzuki (1982)

Table II: Total metal concentrations ($\mu\text{g L}^{-1}$) in pre-melt snowpacks from various remote, North American, and European locations. Data are mean or mid-range of values selected from each reference. "Less than" data in the references were taken at the nominal values when determining means; such means are indicated by "<". Consult text for other data selection criteria.

Location	Cu	Ni	Zn	Pb	Cd	Fe	Mn	Al
Remote Areas								
Arctic ^a	0.034	0.060						
Greenland ^b	0.045	<0.13	<0.27	0.037	0.002			
Antarctica ^c	0.025		0.072	0.106	<0.013	<2.9	<0.035	
				0.028	0.010		0.006	
North America								
Eastern Canada^d								
Ontario ^e	<0.6	2.1		3.2				
Montreal (urban) ^f	28.	43.	94.	<2.7	<0.1	21.	1.5	21.
				8.	6.			
Europe								
S Norway ^g	13.					40.	11.	69.
Norway (S of 65°) ^h	2.1	63.	30.	3.4				
Norway (N of 65°) ^h	4.1	9.6	3.4	0.45		17.		
N Sweden ⁱ	0.38	7.3	2.7	0.43				15.
USSR ^j	11.0	3.1	1.6	0.03				15.
		30.	5.0	0.9		12.	1.	

a Mart (1983), b Davidson et al. (1981), c Boutron (1982), d Barrie and Vet (1984), e Murphy and Robertson (1979), f Landsberger et al. (1982), g Johannessen and Henriksen (1978), h Gjessing et al. (1976), see also Wright and Dovland (1978), i Ross and Granat (1986), j Zoiotareva (1984)

Table III: Concentration of selected pesticides and total PCB's (ng L⁻¹) in snowpack samples from various locations. Sampling and analytical methodologies, and detection limits vary greatly among the studies referenced. Below detection data for those references not specifying detection limits are shown as nd.

Compound*	Ellesmere Island ^a	Canadian Arctic ^b	Ontario ^c	Ontario ^d	FRG ^e	Antarctica ^f
α-BHC	<1-18	0.43-8.72	0.5-1.5			
δ-BHC	<1-8	0.22-4.08	nd-0.4		16-30	
α-chlordane	<1-2	<0.13-0.40		<1	700-1300	1.5-4.9
δ-chlordane	<2-2	<0.13-0.48				
op'-DDT	<1-4					
pp'-DDT	<1-2					
Σ DDT				<1-2		
α-endosulphan	<1-2	<0.13-1.34	0.3-1.9	<1-4		
HEOD	<2-4	<0.13-1.39	nd			0.009-0.016
methoxychlor			nd			
HCB			0.1-5.8	<1-1		
			nd-0.1			
Σ PCB		<0.05-1.67	18-43	<1		
				<10		0.160-1.000

* δ-BHC = lindane, HEOD = dieldrin, HCB = hexchlorobenzene
 a McNeely and Gummer (1984), b Gregor et al. (in press), c Strachan and Huneault (1979), detection limits not specified; d Murphy and Robertson (1979), data presented only when values reported at 2 or more sampling sites; e Schrimppf (1980); f Tanabe et al. (1983)

Table IV: Dry deposition as a percentage of total atmospheric deposition for selected ions in rural areas in N. Michigan (Cadle et al., 1986) and central Ontario (Semkin and Jeffries, in press).

Ion	N. Michigan	Central Ontario
H ⁺	1 - 13	17 - 22
NH ₄ ⁺	14 - 25	4 - 17
Ca ²⁺	19 - 45	5
SO ₄ ²⁻	11 - 30	11 - 13
Cl ⁻	15 - 44	28