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**Ionic elution from the acidic snowpack
during spring thaw period
in the northern part of Hokkaido**

by

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北海道北部における春期融雪時期の酸性積雪からのイオン溶出

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Summary

The chemical composition of the snowfall and the meltwater in the forest area of northern Hokkaido was investigated during the 1995-1996 winter season. The snowfall pH was acidic (4.61, average, volume-weighted mean). The dominant ion species were Na⁺ and Cl⁻, indicating the strong influence of sea salt.

A marked depression of pH values, dropping below 4.0, was obtained in the initial stage of the spring thaw, and the H⁺ concentration decreased rapidly with the progression of the thaw. Approximately 70% of H⁺ eluted out during the thaw was lost with the first 20% of meltwater. However, the concentration of other major ions did not decrease even in the later stage of the thaw.

The amount of ions eluted in the snowmelt exceeded the total ion loading by snowfall. The influence of dry deposition on the chemical characteristics of the snowpack was also estimated from the ionic composition of the snowmelt.

Key Words : acid snow, chemical fractionation, dry deposition, spring melt, acid shock

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1. Introduction

The chemical composition of meltwater and its influence on forest ecosystems has been extensively studied during the last two decades in the cold and snowy regions of Europe and North America (*e. g.* Johannessen and Henriksen, 1978, Morris and Thomas, 1985, Tranter, *et al.*, 1986, Williams and Mellack, 1991, Jenkins *et al.*, 1993). These studies have shown that the sudden release of acid materials from accumulated snow during the early spring thaw (chemical fractionation: Tranter *et al.*, 1987) results in the rapid, temporary acidification of stream and lakes causing severe damage on the aquatic biota (acid shock).

Hokkaido Island, Japan, is located at the northeastern margin of the Far East. About 30~40% of the annual precipitation is supplied as snowfall, most accumulating on the ground as snowpack, in the forest watersheds. As the concentration of acidic pollutants in the snowfall of this region is roughly the same degree as that observed in Europe and North America (Sato *et al.*, 1996a), strong elution of ions (including H^+), which affects stream chemistry, is expected to take place through chemical fractionation process.

On the other hand, acidic pollutants in snowpack accumulate in the form of not only wet deposition (snowfall) but also dry deposition. Our investigations suggest that dry deposition affects the chemical composition of newly fallen snow (Sato *et al.*, 1996b, 1996c), although the contribution of dry deposition to the chemical characteristics of snowpack is generally considered to be smaller than that of wet deposition (Cadle, *et al.*, 1984, Jeffries, 1990). It is therefore also interesting to evaluate the contribution of such deposits quantitatively.

This paper reports the results of the study on the chemistry of snowfall and the snowmelt in northern Hokkaido during the 1994-1995 winter season (December 1994 to April 1995). A discussion of chemical fractionation and the significance of dry deposition are also included.

2. Study area and methods

The sampling of snowfall and meltwater was carried out in an open field near the station of Teshio Experimental Forest (44° 54' 46" N, 142° 1' 40" E, Fig. 1), located about 70km south of Wakkanai, the northernmost city in Japan. As the experimental site was surrounded with mixed forests and pasture lands, there was no industrial source of acidic pollutants within 70km (Wakkanai city) of the site. Meteorological observations were also carried out at this site.

Daily observation of newly fallen snow was performed at 9 a.m. throughout the winter season. At the same time, a snowfall sample was taken when an accumulation of newly fallen snow was recognized on the sampling board (50cm×50cm). All snow on the board was taken for the investigation of its amount and chemical composition. Snowmelt samples were collected with following procedure. A stainless lysimeter (100cm×100cm×20cm) was placed on the ground early in December before the development of continuous snowcover. The meltwater from the bottom of the snowpack was stored temporarily in the polyethylene reservoir installed in the ground so as to avoid freezing. The water sample in the reservoir was then sucked up to the ground surface by use of a hand-operated vacuum pump. This sampling was undertaken once in a day (at 9 a.m.) until the beginning of spring snowmelt and then twice (9 a.m. and 5 p.m.) during spring thaw period.

The snowfall sample was transferred to the laboratory and then melted at room temperature. The snowfall and snowmelt samples were stored in a refrigerator at 4°C after the determination of

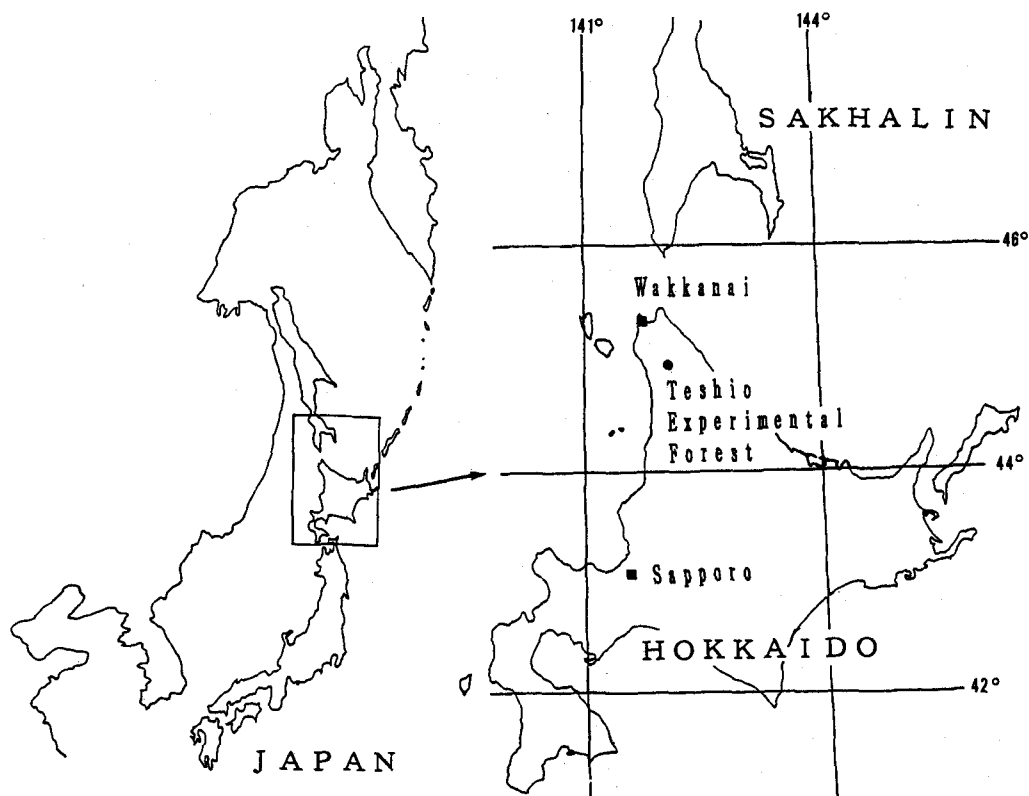


Fig. 1. Location of experimental site.

pH and electric conductivity (EC). Analysis of the major ions was performed by ion chromatography on a TOA Model 3000 System after the filtration with a $0.22\ \mu\text{m}$ membrane filter.

3. Results and discussion

3.1. Winter climate

Fig. 2 gives the maximum and minimum daily temperature, thickness of the snowpack, cumulative snowfall and cumulative snowmelt at the experimental site during 1994~1995 winter. The minimum daily temperature in period December~March typically stayed below zero and sometimes dropped to near -30°C . The maximum daily temperature, however, fluctuated around 0°C and often rose close to 10°C . Such short periods of warm temperature occasionally appear during the normal winter in this area. Approximately 450mm water equivalent precipitated as snowfall. The snowcover immediately developed thickness of more than 50cm early in December, which was sufficient to prevent the ground becoming frozen in this area (Sasa, 1993). The snowcover maintained this thickness until mid-April just before the disappearance of the snowpack.

Since the ground did not freeze, successive melt water appeared late in December. Motoyama (1986) has reported this snowmelt in mid-winter and termed it "bottommelt". The bottommelt continued for more than 100 days during winter until the occurrence of spring snowmelt, mid-March. The cumulative amount of the bottommelt accounted for about 13% of total water equivalent as snowfall, although daily water equivalent as bottommelt was less than 1 mm.

Snowmelt proceeded rapidly from mid-March (spring melt), when the minimum daily temperature rose to 0 °C and the maximum daily temperature exceeded 10 °C. The amount of daily snowmelt was around 10mm/day and increased to 20~40mm/day when spring rain occurred. The amount of spring snowmelt (including spring rain) was 450 mm and the total amount of the snowmelt in this season (springmelt plus bottommelt) was 504 mm.

3.2. Snowfall (wet deposition)

The average pH of the snowfall was 4.61 (volume-weighted mean, Table 1) and the total loading of H⁺ was about 0.10 kmolc/ha (Table 2), which was about the same order as those reported in the forest areas of Europe and North America.

The total loading of cations and anions by snowfall was 1.57kmolc/ha and 1.48kmolc/ha, respectively (Table 2). Na⁺ and Cl⁻ were the dominant ion species in the snowfall, since the surveyed area was located only about 20km from the coast of the Sea of Japan. Under the assumption that all Na⁺ and Cl⁻ originated from the sea salt, magnesium and sulfate sea salt-related fraction can be assessed. The snowfall loading of Mg²⁺ and

SO₄²⁻ originating from sea salt was estimated to be 0.25kmolc/ha (Mg²⁺/Na⁺=0.23: Ball *et al.*, 1980) and 0.13kmolc/ha (SO₄²⁻/Cl⁻=0.11), respectively. The total loading, however, was 0.27kmolc/ha for Mg²⁺ and 0.24kmolc/ha, which suggests that nearly 50% of the SO₄²⁻ loading was in non-sea salt form. On the basis of observations on the chemical composition of the snowfall for five years at this site, we suggested that the source of the non-sea salt SO₄²⁻ (excess sulfate)

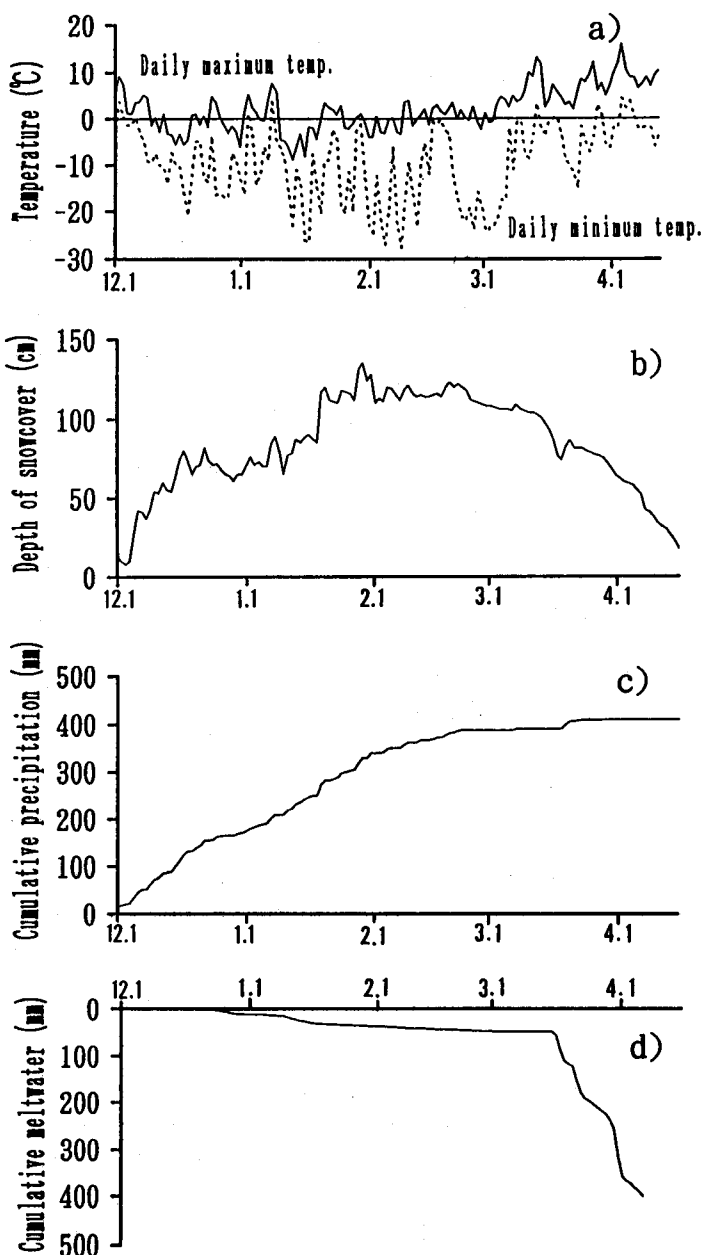


Fig. 2. Daily maximum and minimum temperature, depth of snowcover, cumulative snowfall (water equivalent) and cumulative meltwater during the 1994~1995 winter.

Table 1

Average pH and average individual components in $\mu\text{molc/L}$ in the snowfall, the bottmmelt and the spring melt in the 1994~1995 winter (weighted mean)

	Snowfall	Bottmmelt ¹⁾	Springmelt ²⁾
pH	4.61	4.32	4.95
H ⁺	24.7	47.3	11.1
Na ⁺	260.6	340.4	370.7
NH ₄ ⁺	33.1	96.4	78.7
K ⁺	9.0	13.6	16.2
Mg ²⁺	77.9	95.4	68.6
Ca ²⁺	13.0	77.3	45.4
Cl ⁻	263.8	373.8	373.3
NO ₃ ⁻	10.9	68.7	28.7
SO ₄ ²⁻	89.3	182.4	101.1

1) melt water until the beginning of spring thaw.

2) melt water during spring thaw.

Table 2

Mass balance for input from snowfall and snowmelt output, and the content of snowmelt (bottmmelt plus springmelt) 1994~1995 winter.

Units are in kmolc/ha unless otherwise specified.

	Input Snowfall	Output snowmelt ¹⁾	bottmmelt ²⁾	springmelt ³⁾
Water equivalent (mm)	448	504	54	450
H ⁺	0.104	0.068	0.023	0.045
Na ⁺	1.087	1.614	0.163	1.451
NH ₄ ⁺	0.143	0.372	0.046	0.326
K ⁺	0.039	0.070	0.007	0.063
Mg ²⁺	0.267	0.312	0.037	0.275
Ca ²⁺	0.028	0.220	0.046	0.174
Cl ⁻	1.191	1.731	0.180	1.551
NO ₃ ⁻	0.050	0.167	0.033	0.167
SO ₄ ²⁻	0.241	0.511	0.088	0.423

1) melt water throughout 1994~1995 winter.

2) melt water until the beginning of spring thaw.

3) melt water during spring thaw.

probably was long range transported materials from mainland China (Satoh *et al.*, 1996a).

Apart from the sea salt substances, a relatively large amount of NH₄⁺ (0.14kmolc/ha) was also detected in the snowfall. The NH₄⁺ loading was about twice that reported in the forest area in Canada (Semkin and Jeffries, 1988). As dairy farming is one of the main activities in northern Hokkaido, contamination by waste from livestock was probably the source of this NH₄⁺.

3.3. Snowmelt

The amount of the bottmmelt, snowmelt during the thaw period (springmelt) and the sum of snowmelt throughout the winter (total melt) are reported in Table 2. About 10% of water equivalent

was lost from the snowpack by the bottommelt until the beginning of the spring thaw. At the same time, from 9% (for Mg^{2+}) to 33% (for H^+) of the ions in the snowpack also eluted out due to the bottommelt. A detailed description and the discussion of the chemistry of the bottom melt have been reported elsewhere (Sato, *et al.*, 1997). The assumption that the loading of chemical substances within the snowpack is conservative, which is usually accepted in high-latitude cold and snowy regions is not valid when the occurrence of bottommelt is evident. In this study, therefore, we adopted the results of the lysimetric study to estimate the chemical loading within the snowpack in stead of the snow core sampling from snowpack just before melting occurred.

The concentration of ionic species eluted from the snowpack during the spring thaw period is shown in Fig. 3. The maximum concentration of H^+ was observed in the initial stage of the spring thaw, when the pH was 3.97, and subsequently decreased rapidly during the progression of the snowmelt. Rainfall in this period has been reported to accelerate the elution of pollutants from the snowpack (Cadle *et al.*, 1984). In spite of the rainfall (on 18, 22 and 31 March), no increase in the concentration of H^+ was observed in the middle of spring thaw. This implies that the elution of H^+ mostly completed prior to the disappearance of the snowpack.

Contrary to H^+ , the concentration of Na^+ and Cl^- was low in the first increment of meltwater by rainfall on 18 March. High elution of both ions, however, was noted in the second (on 22 March) and third (on 31 March) increments. The elution of Mg^{2+} and SO_4^{2-} had similar tendency to that of Na^+ and Cl^- . This elution pattern indicates the contribution of freeze and thaw process within the snowpack.

In high latitude region, a pronounced elution of ions has been observed in the first melt. The segregation of chemical substances at the grain surface of the snow and the presence of freeze-thaw

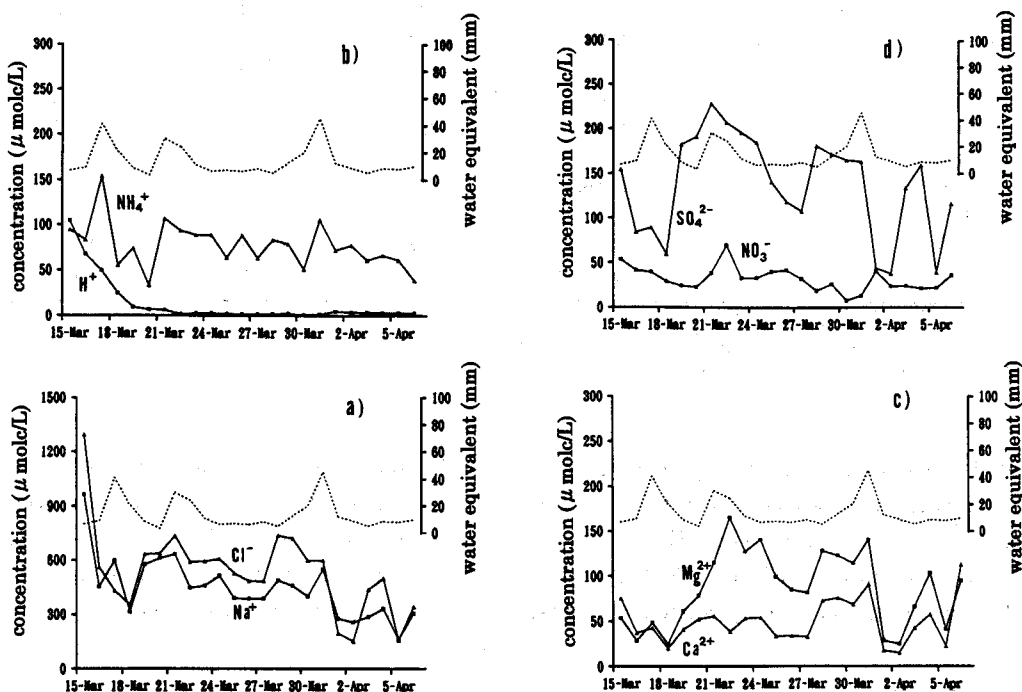


Fig. 3. Concentrations of major components and melt water equivalent during the 1995 thaw period. Dashed line indicates melt water equivalent.

events are considered to be important in this phenomenon (Jones, 1985). As shown in Fig. 2, however, strong elution of ions (except H^+) was not found in the early melt of spring thaw.

The chemical fractionation of ions released during the thaw period was estimated from the amount of melt water and the ion loading in snowmelt (Fig. 4).

About 70% of total H^+ was exported from the snowpack with the first 20% of the melt water. As for other ions, however, 50~60% of the total ion loading in the melt water corresponded to 40~50% of the melt water. This indicates that the concentration of ions in initial spring meltwater was not so strong as that reported in north Europe and America (e.g. Brimblecombe, *et al.*, 1985). In temperate areas like northern Hokkaido, the segregation process does not proceed to an extent that can cause a strong concentration of impurities.

The occurrence of bottommelt and the occasional warm period is likely to prevent re-crystallization of the snow at the bottom of the snowpack and the subsequent repetition of freeze-thaw events.

3.4. Dry deposition

Dry deposition may occur on the snowpack and increase the chemical load in snowmelt. The amount of dry deposition was estimated by comparing the cumulative wet deposition (calculated from snowfall in winter and rainfall during thaw period) with the cumulative snowmelt from this lysimetric study (Fig. 5). The total water equivalent of snowmelt during whole winter season was 504mm (Table 2), which was higher than that of precipitation (448mm, as snowfall in winter and rainfall in spring thaw). Excess meltwater from surrounding snowpack was considered to result in such a difference. Nomura (1994) indicated that the development of the rout of meltwater was not uniform within snowpack according to the formation of ice plates. The snowpack of investigated site included many ice plates, so it is reasonable that lateral movement of meltwater took place and poured into the lysimeter from its outside area of snowpack. As the difference of water equivalent between precipitation and snowmelt was about 10%, we adopted the result of the lysimetric study to estimate the amount of dry deposition.

As shown in Table 2, the amount of ions, except for H^+ , eluted from the snowpack exceeded ion

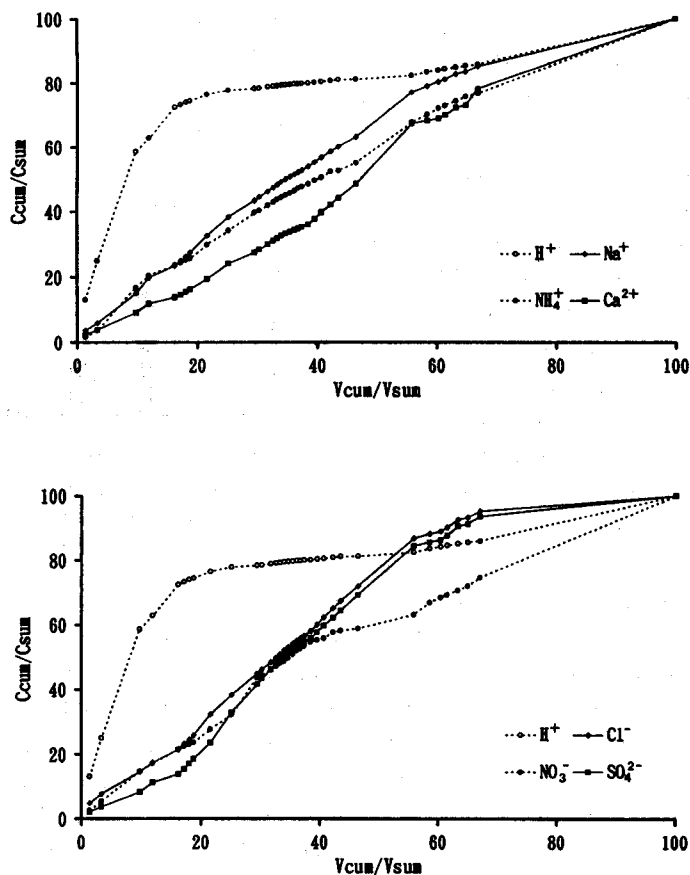


Fig. 4. Percent loss of major components as a function of percent water loss the snowpack during the 1995 thaw period.

loading through precipitation. The rate of dry deposition ranged from 31% (for Cl^-) to 87% (for Ca^{2+}) of total mass balance. Especially, concerning to NH_4^+ , Ca^{2+} , NO_3^- and SO_4^{2-} , which are correlated with anthropogenic pollutants, the contribution of dry deposition accounted for more than 50%. As mentioned above, however, it includes some over-estimation as to the amount of dry deposition. Even though such uncertainty is taking into account, it can be said dry deposition is significant for the winter deposition of this area.

The contribution of dry deposition to snowmelt loading has been considered to be smaller than wet deposition (Jeffries, 1990). Our snowmelt study, however, implied dry deposition to be as important as was wet deposition in the northern part of Hokkaido. Dry deposition affects the chemical composition of the snowpack in this area due to the adsorption on the surface of the snowpack (Satoh et al., 1996b). The amount of H^+ in the snowpack in the forest can be much lower than in the open field near the forest of this area (Satoh et al., 1996c). This reduction of acidity in the snowpack is caused by the reaction of H^+ included in snowpack with dry deposition, especially for NH_4^+ , trapped by leaves, branches and trunks (Jones and Sohanska, 1985, Sakuma, 1994). These findings strongly support the contribution of dry deposition to the chemical composition of the melt water. In winter, strong westerly winds usually blow over this area, so that dry deposition is probably transported continuously from the Sea of Japan and the mainland China under the strong wind conditions, whenever snowfall does not take place.

4. Conclusion

Northern Hokkaido is a representative area of the cold and snowy region in Japan. Compared with high-latitude cold and snowy regions, however, the more temperate winter climate of this study area prevents soil freezing and frequently causes bottommelt throughout mid-winter. Under such conditions, a substantial loss of ions through the bottommelt results in weak chemical fractionation (except for H^+). Approximately 30% of H^+ loaded through snowfall has already been lost by bottommelt, which suggests that the acidity of snowpack was reduced due to bottommelt.

The deposition of acid pollutants is striking in the forest watersheds of northern Hokkaido. The snowpack containing acid substances considered inducing 'acid shock' to stream water. A reduction in the acidity of the snowpack would favour stream water ecosystems. Despite the fact that these observations were performed in an open field, dry deposition had some effect reducing the acidity of the snowpack of investigated area. When the snowpack is surrounded by many trees that trap dry deposition, the acidity of the snowpack is probably far less than that in open locations.

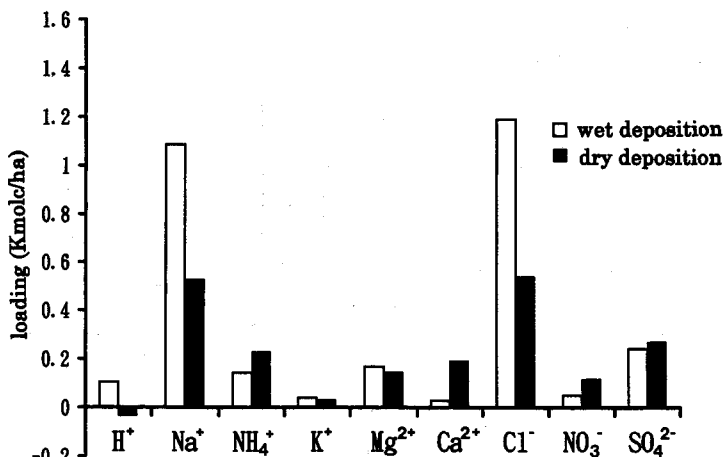


Fig. 5. Loading by wet deposition (snowfall) and dry deposition estimated from the difference between input by the snowfall and output by the snowmelt during the 1994~1995 winter.

Consequently, more detailed studies are necessary to clarify the effects of forests on the chemistry of snowpack and preferential elution in this area.

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要 旨

北海道北部森林地帯における降雪と融雪水の化学組成について、1995-1996年冬期間に調査をおこなった。降雪pHは酸性であり（加重平均で4.61）、イオン組成は海塩の強い影響を受けて Na^+ と Cl^- が主体であった。

春期融雪初期の融雪水pHは4.0以下の著しく低い値を示したが、融雪水中の H^+ 濃度は融雪の進行するにつれて急激に低下した。融雪期に溶出した H^+ の約70%が初期の融雪水（春期融雪水中の約20%）とともに積雪より失われた。しかし、他の主要イオンは融雪後半になっても濃度低下はあまり顕著ではなかった。

融雪水とともに溶出したイオン総量は降雪によるイオンの付加量を上まわっており、乾性降下物の積雪の化学組成への影響が推定された。

キーワード： 酸性雪，化学的分画，乾性降下物，春期融雪，アシッドショック