

# Acid precipitation in Norway: the regional distribution of contaminants in snow and the chemical concentration processes during snowmelt

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**Abstract.** Regional surveys of snow conducted in late winter 1974 and 1975 as part of the SNSF-project (Acid precipitation—Effects on forest and fish) show that southeastern Norway receives massive deposition of atmospheric pollutants such as  $H^+$ ,  $SO_4^-$ ,  $NO_3^-$  and several heavy metals. Polluted snow was also found in 1975 areas of northernmost Norway.

Laboratory and field experiments demonstrate that the concentration of these pollutants is 2–3 times higher in the first meltwater fractions than in the bulk snow. Subjecting the snow to melt-freeze cycles enhances this concentration effect. The melting of contaminated snow causes sudden drops in pH and produces drastic effects on fresh-water ecosystems in Norway.

**Les effets de la précipitation acide en Norvège: la distribution régionale des polluants de neige et les processus de concentration chimique pendant la fonte de la neige**

**Résumé.** Recherches régionales sur des échantillons de neige collectée tard en hiver 1974 et 1975 effectuées comme une partie d'un projet SNSF (Les effets de la précipitation acide sur les forêts et poissons) montrent que le sud-est de la Norvège reçoit des dépositions massives de polluants tels que  $H^+$ ,  $SO_4^-$ ,  $NO_3^-$  et métaux lourds. En 1975 on a également trouvé de la neige polluée en régions étendues de la Norvège la plus boréale.

Des expériences en laboratoire et dans le terrain démontrent que la concentration de ces polluants est 2 à 3 fois plus haute dans l'eau au début de fusion que dans la totalité de l'eau de la neige fondue. Cette concentration augmente quand on fait fondre et geler successivement la neige. Lorsque la neige polluée fond, cela provoque des chutes soudaines du pH résultantes des effets radiaux sur les systèmes écologiques de l'eau douce en Norvège.

## INTRODUCTION

Air pollutants originating in Great Britain and central Europe are transported over long distances and produce highly polluted precipitation over large areas of Scandinavia (Bolin *et al.*, 1971; Brosset, 1973; Førland, 1973; Rühling and Tyler, 1973). In Norway a considerable fraction of the precipitation falls as snow, and during recent years grey snow has frequently been observed (Henriksen, 1972; Elgmork *et al.*, 1973; Dale *et al.*, 1975).

The Norwegian project entitled *Acid Precipitation—Effects on Forest and Fish* (SNSF-project) was initiated in 1972 to document the amount and the effects of atmospheric pollutants in Norway (Overrein and Abrahamson, 1975). Measurements of the contaminant load in the snowpack and the release of these pollutants during snowmelt forms a major part of the project.

## REGIONAL SURVEY OF SNOW CHEMISTRY

In late winter 1974 samples of the snowpack were collected from 24 localities in Norway and the concentration of  $H^+$ , Mg, Ca, Na, K,  $SO_4$ ,  $NO_3$ ,  $NH_4$ , total-P, ortho-P, Cl, Cu, Zn, Cd and Pb were determined (Dale *et al.*, 1975).

The results show that the deposition of the pollutants  $H^+$ ,  $NO_3$ , excess- $SO_4$  (corrected for sea-water sulphate),  $NH_4$  and Pb, is largest in southernmost Norway and decreases to the north and the northwest (Fig. 1).

The snow survey was repeated in 1975 and expanded to include duplicate samples taken at 80 localities distributed all over Norway. Figure 1 shows that

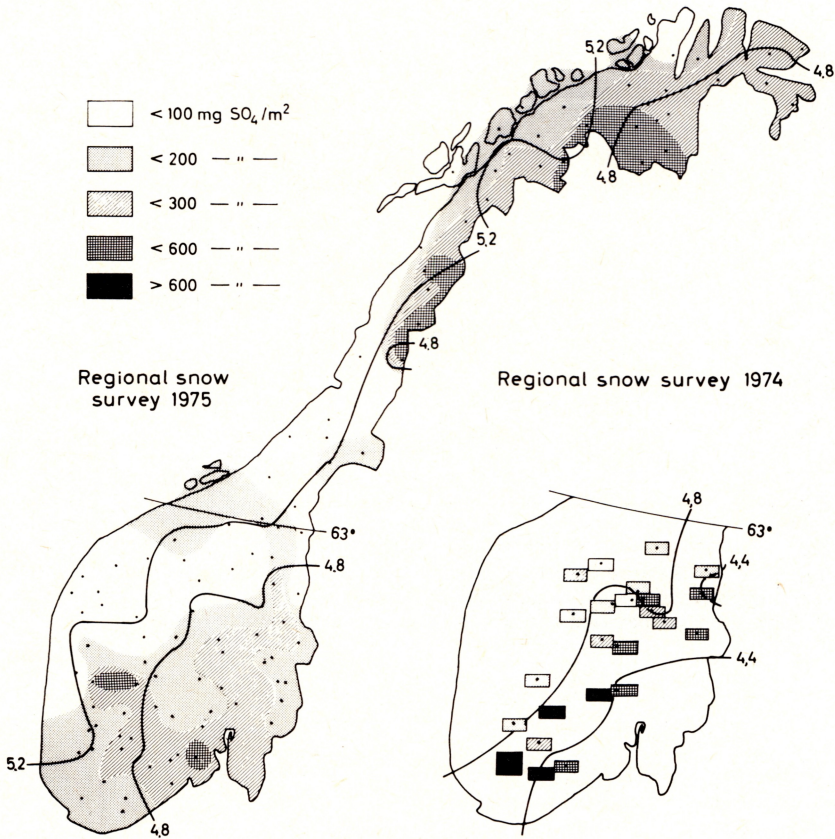


FIGURE 1. Excess-SO<sub>4</sub> (mg/m<sup>2</sup> corrected for sea-water sulphate) and pH.

the deposition of excess-SO<sub>4</sub> (mg/m<sup>2</sup>) and pH of the samples again gave high pollutant loads in southeastern Norway although at lower levels than the year before. In addition, high levels of acid components were found in the snowpack in northernmost Norway.

The deposition of these pollutants is often episodic, and the pollutant load of a given episode is apparently dependent on the storm track (Førland, 1973). Indeed snow falling during such episodes is often grey and marker-horizons can be found in snow over large areas (Dale *et al.*, 1975).

RELEASE OF POLLUTANTS DURING SNOWMELT

The pollutants accumulated during the winter are released during a short period in the spring. The snowmelt process is thus critically important in the evaluation of the impact of contaminants on freshwater ecosystems.

The release of chemical components from polluted snow collected in southeastern Norway was investigated using a plastic pipe 56 cm long and 13 cm in diameter with a sintered disk fitted to the bottom end. During melting the temperature of the lysimeter was regulated at 2.5–3°C and under a 1.1-atm. pressure of N<sub>2</sub> the meltwater was drawn off through an outlet at the bottom and 25-cm<sup>3</sup> aliquots collected with a fraction collector.

Figure 2 shows a typical melt curve where the chemical concentrations are expressed in terms of the relative concentration in the aliquots *c*, to the

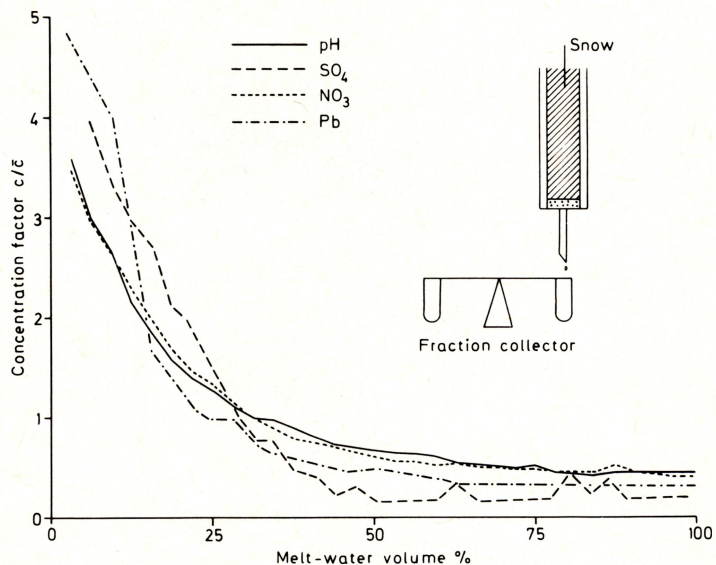


FIGURE 2. Relative concentration of  $H^+$  (as pH),  $SO_4$ ,  $NO_3$ , and Pb (concentration in the meltwater fraction  $c$ /concentration in the bulk snow  $\bar{c}$ ) in meltwater fractions from a polluted snow sample. *Insert*: schematic diagram of laboratory lysimeter.

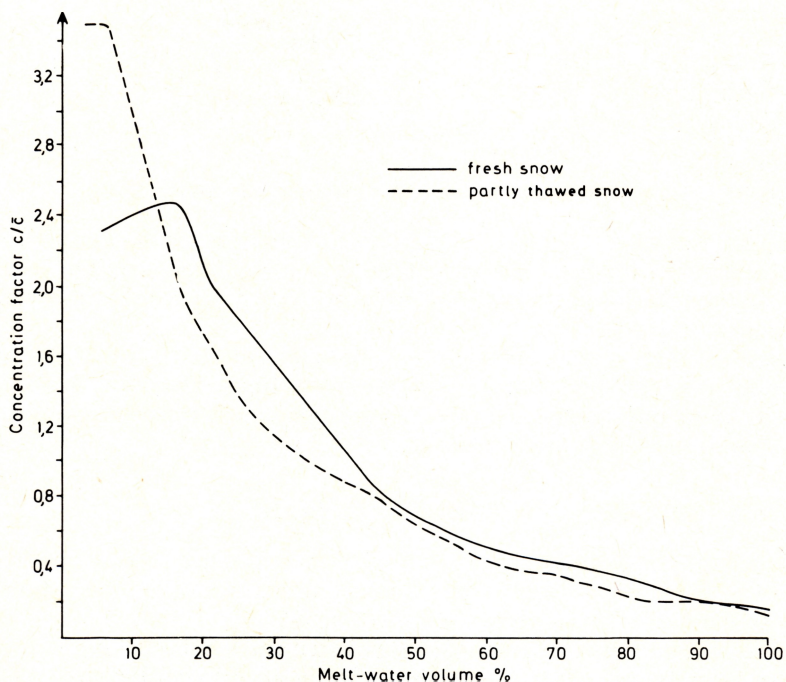


FIGURE 3. Relative pH (pH in meltwater fraction  $c$ /pH in bulk snow  $\bar{c}$ ) in meltwater fractions from a fresh snow sample and a partly thawed sample.

concentration in the bulk snow  $\bar{c}$ . The concentration of eight chemical components are 3–5 times higher in the first fractions than in the bulk snow. The fraction of each component released in the melting of the first 30 per cent

of the snow sample is listed in Table 1. The results are similar to those reported by Odén and Bergholm (1972).

Laboratory experiments designed to approximate the melt-freeze-melt cycles that occur during natural snowmelt demonstrate an even greater concentration effect. Figure 3 shows the results for pH from the simple melting of a fresh snow sample and from the same snow subjected first to a slight thawing and refreezing. The partial thawing and refreezing produces an increase in the amount of chemicals (especially the divalent ions  $\text{SO}_4$ , Ca and Mg) released in the first 30 per cent of the meltwater (Table 1).

TABLE 1. Percentage of each component released in the melting of the first 30 per cent of the snow sample

	H <sup>+</sup>	SO <sub>4</sub>	NO <sub>3</sub>	NH <sub>4</sub>	Na	K	Ca	Mg	Cl	Mn	Zn	Cu	Pb	Cd	Cr
Polluted snow	64	76	63		67		71	90					83	31	
Fresh snow	63	59	63	52	54	58	64	61	64	63	66	64	75	48	54
Partly thawed snow	68	71	64	53	54	50	78	73	58	80	73	60	68	54	62

The effect of melt-freeze cycles under natural melting were confirmed by results from a field lysimeter in the winter of 1973–1974. This field lysimeter, shown in Fig. 4, is a polyethylene cylinder 1 m in diameter and 40 cm deep that is buried in the ground such that the top extends about 3 cm above the surface and is covered by a perforated polyethylene sheet. Over the winter the snowpack accumulates naturally on top of the lysimeter, and during the periods of melting the meltwater runs into the cylinder and is collected in a polyethylene bottle.

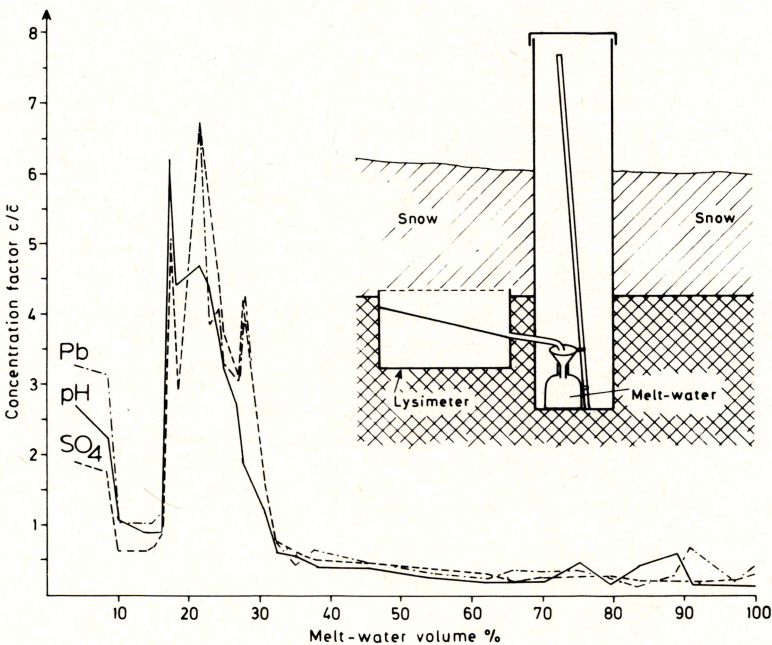


FIGURE 4. Relative concentrations of H<sup>+</sup> (as pH), SO<sub>4</sub> and Pb (concentration in meltwater fraction *c*/concentration in bulk snow  $\bar{c}$ ) in meltwater fractions collected under natural snowmelt conditions. The initial concentration drop in the first 15 per cent of the meltwater was due to an early-winter mild period in which the early snowfall melted. *Insert*: schematic diagram of field lysimeter installation.

The results from the natural melting follow the pattern of the laboratory experiments, but the concentration effect is even greater. Some of the earliest meltwater fractions contain about 5–6 times the concentration in the bulk snow, and the first 30 per cent of the meltwater contains 70–80 per cent of the total amount of  $H^+$ ,  $NO_3$ ,  $SO_4$ , Cl, Ca, Mg, Na and K (Table 1).

## IMPACT ON FRESH-WATER ECOSYSTEMS

The pollutants released during the first phase of snowmelt cause sudden changes in water quality of streams and lakes. During the spring, pH-drops of more than 1 unit cause severe physiological stress to fish and other aquatic organisms and occasionally result in massive fish kills (Hagen and Langeland, 1973; Muniz *et al.*, 1975). Rapid changes in acidity may be the primary impetus for the 'self-accelerating oligotrophication' proposed by Grahn *et al.* (1974) and Hultberg (1975).

The accumulation of pollutants in the snowpack over the winter, and the sudden release of high concentrations of those pollutants during the first phases of snowmelt are the major factors in the acidification of Norwegian fresh-waters.

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