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Episodic acidification in northern Sweden: a regional assessment of the anthropogenic component

H. Laudon[a,*], O. Westling[c], A. Bergquist[d], K. Bishop[b]

[a]Department of Forest Ecology, Swedish University of Agricultural Sciences, SE-901 83 Umeå, Sweden
[b]Department of Environmental Assessment, Swedish University Agricultural Sciences, SE-750 07 Uppsala, Sweden
[c]Swedish Environmental Research Institute, Aneboda, SE-360 30 Lammhult, Sweden
[d]Department of Ecology and Environmental Science, Umeå University, SE-901 87 Umeå, Sweden

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Abstract

Spring flood is the period in boreal ecosystems that is most sensitive to acid deposition since as much as half of the year's precipitation melts and enters streams or the soil in the space of a few weeks. The 'Episode Project' in northern Sweden found a consistent relationship between the SO_4^{2-} concentration of snow and the anthropogenic component of acid neutralization capacity (ANC) decline during spring flood. This correlation creates the possibility for a regional prediction of the severity of anthropogenic episodic acidification during spring flood episodes using SO_4^{2-} deposition data together with chemical data from 1240 lakes selected to be representative of northern Sweden. The regional assessment found that in 1998, ca 6% of the region was seriously affected by anthropogenic acidification during spring flood. The results from this study have important implications for both the national liming strategy and international negotiations to further reduce emissions of air pollutants in Europe.

Keywords: Episodic acidification; Regionalization; Anthropogenic deposition; Spring flood; Northern Sweden

1. Introduction

Episodic acidification associated with rainfall and snowmelt constitutes an important water quality threat in large regions of the world (Wigington et al., 1992; Davies et al., 1992). Models to better differentiate driving mechanisms of episodic acidification using high resolution time series of water chemistry have greatly improved our understanding of the role of acid deposition in individual streams (Molot et al., 1989; Wigington et al., 1996; Laudon and Bishop, 1999). But in order to synthesize results from individual episodes to regional assessments, new models are needed which can make use of geographically available data with limited temporal resolution. One example of a regional model is the 'index approach' developed by Eshleman (1988) and later used in a landscape scale study in the Northeastern USA to predict minimum acid neutralization capacity (ANC) associated with spring melt runoff (Eshleman et al., 1995). Similar approaches based on the baseflow

* Corresponding author. Fax: +46-9078-677-50.
E-mail address: hjalmar.laudon@sek.slu.se (H. Laudon).

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chemistry of surface waters to estimate the most acid conditions during rain driven episodes have subsequently been adopted by both Gerritsen et al. (1996) and Davies et al. (1999), but they also incorporated either precipitation information or change in discharge as independent variables.

In a large multi-investigator project in northern Sweden, the Boreal Dilution Model (BDM) was developed to quantitatively distinguish the natural and anthropogenic mechanisms that drive episodic ANC and pH decline during hydrological events (Bishop et al. 2000; Laudon, 2000). Northern Sweden is a region where the anthropogenic deposition component in episodic pH and ANC decline has generated considerable interest and controversy during the last decade (Bishop, 1997; Bishop et al. 2001). Despite a relatively low acid deposition load (4–8 kg S ha^{-1} yr^{-1} during the 1980's and 2–4 kg S ha^{-1} yr^{-1} during the 1990's) many surface waters in the region experience low ANC and pH in conjunction with hydrological episodes, especially during spring melt events when as much as 50% of the annual runoff from small headwater catchments can occur. Since there has been concern that anthropogenic deposition in the region during spring runoff threatens the natural biota, the Swedish government has subsidized liming in northern Sweden with approximately one billion Swedish crowns (125 million US$) during the last decade. A continuation of that liming in the coming decade is planned despite the halving of acid deposition levels in the region during the last two decades (Swedish Environmental Protection Agency, 1999).

Information provided by the BDM on natural and anthropogenic components of episodic pH decline can help assess the need for further liming to remediate surface water acidification. In order to run the BDM, though, a time series of water chemistry data from hydrological episodes are necessary. Because such data are not commonly available, a 'one point' version of the BDM (pBDM) was presented by Laudon (2000), which can be applied using the much more widely available data on water chemistry in lakes or stream baseflow. The pBDM is based on the consistent relationship between the SO_4^{2-} content of snow and the anthropogenic component of ANC decline during spring flood together with a relatively predictable hydrology during spring melt. These empirical relationships discovered in several dozen applications of the BDM lay the basis for the model proposed here to provide a synoptic overview of human impact on spring melt ANC in northern Sweden using widely available lake chemistry measurements. The concept presented here is similar to the 'index approach' presented by Eshleman et al. (1995) in that it is based on baseflow chemistry to predict the decline in ANC. The key difference is that this model predicts both the natural component in ANC decline during spring runoff as well as the extra acidity caused by acid deposition.

2. Data

The predictive model presented here is based on 53 spring flood episodes from northern Sweden collected during the 1990's (Laudon and Bishop, 1999; Laudon et al. 2000; Laudon and Hemond, 2002). The model is applied to 1240 small head water lakes (Table 1) sampled during the winter together with SO_4^{2-} concentrations in snow (bulk precipitation) during the 1997/1998 winter from the Swedish Precipitation Chemistry Network (Kindbom et al, 2000).

Since northern Sweden is a heterogeneous region with regards to both geological history and SO_4^{2-} deposition, the region is divided into six physiographic sub-regions in order to visualize the more local pattern

Table 1
Source of lake data

N lakes	Data set	Lab	Source
1103	National lake inventory 1990	1	Bernes, 1991 (Monitor 12)
44	National reference lakes	1	Wilander, 1997 (SNV 4652)
43	Boreal lake survey	2	Bishop (1995)
14	MSc Thesis	2	Dahlström and Johansson (1997)
36	Unpublished material	2	A. Bergquist (Unpublished)

Anions were analyzed using liquid ion chromatography analysis and base cations using ICP-MS at the Department of Environmental Assessment, Swedish University of Agricultural Sciences (SLU), Uppsala, (Lab. 1) and SLU Environmental Quality laboratory, Umeå (Lab. 2).

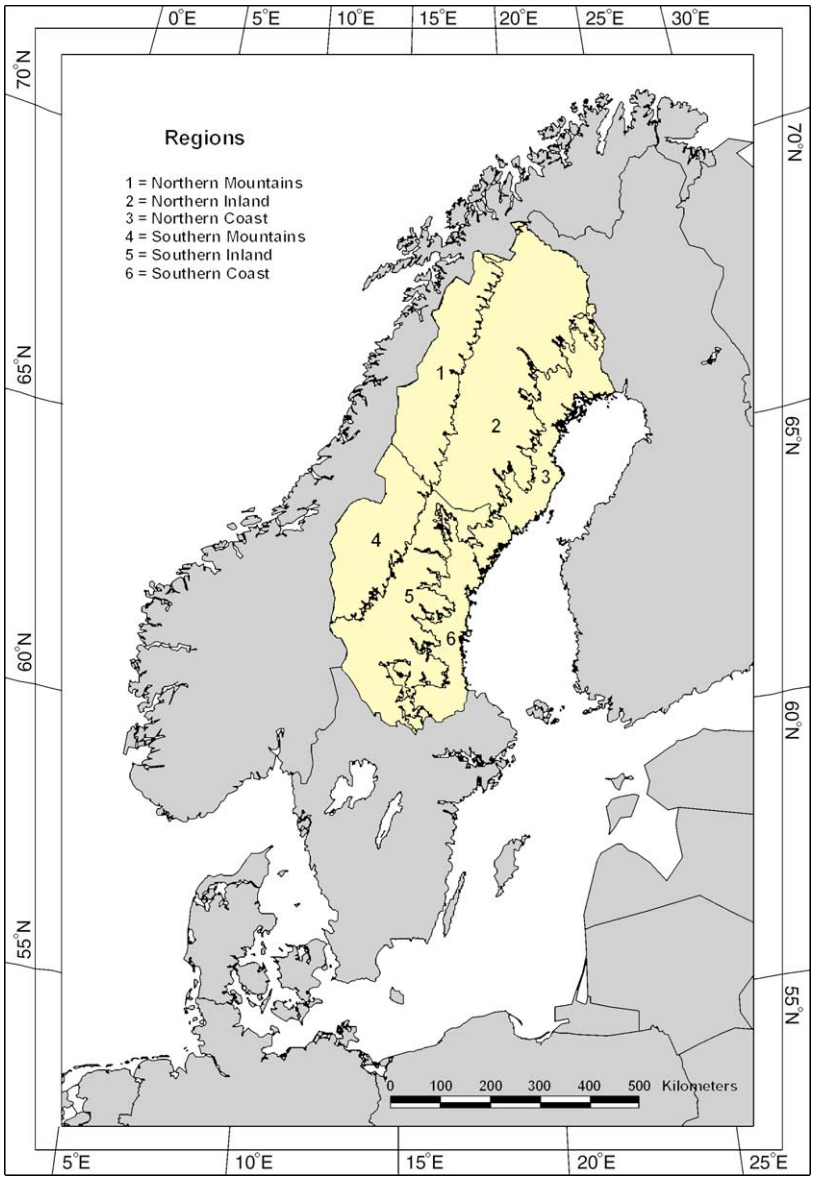

Fig. 1. Map over northern Sweden with sub-regions.

of spring flood response to acid deposition (Fig. 1). In the East-West direction the regions are separated into sub-regions with distinct geological history. The sub-regions along Sweden's western border to Norway are part of the Caledonian mountain range (1 and 4) that consists mainly of folded sedimentary bedrock. The most Eastern sub-regions (3 and 6) along the Baltic Sea coast are below the highest post-glacial coastline (which, depending on latitude, is 160–260 m above sea level). The subregions in-between the Caledonian mountain range and the coastal area are the Inland sub-regions (2 and 5). The coastal and Inland sub-regions are situated on the pre-Cambrian shield and consists mainly of gneisses along the coast and coarse grained granites inland. In the North to South direction, the division into the northern sub-regions (1–3) and the southern sub-regions (4–6) is at the administrative border between the two most

Table 2
Sub-region lake characteristics

Region	Number of lakes	Meters above sea level	Lake size (km^2)[a]	SO42- ($\mu eq\ l^{-1}$)	BC ($\mu eq\ l^{-1}$)	ANC ($\mu eq\ l^{-1}$)	F-factor	SO4 dep 1997/1998 ($\mu eq\ l^{-1}$)
S. Coast	172	149 (76)	0.9	112 (51)	355 (158)	202 (141)	0.91 (0.13)	28 (6)
S. Inland	349	344 (104)	<0.1	62 (32)	330 (243)	218 (180)	0.81 (0.20)	21 (4)
S. Mountains	65	559 (181)	<0.1	53 (36)	315 (219)	233 (203)	0.78 (0.27)	18 (5)
N. Coast	127	111 (68)	0.6	93 (60)	353 (151)	221 (129)	0.89 (0.13)	27 (5)
N. Inland	401	361 (98)	0.2	43 (24)	284 (166)	225 (174)	0.77 (0.22)	14 (5)
N. Mountains	126	657 (197)	0.4	56 (29)	348 (233)	249 (191)	0.81 (0.24)	11 (1)

Numbers in parentheses are standard deviations.
[a] Calculated as the median.

northern counties in Sweden (Västerbotten and Norrbotten) and the more southern counties (Jämtland, Västernorrland, Dalarna and Gävleborgslän). More characteristics of the regions are presented in Table 2.

3. Predicting regional episodic acidification in northern Sweden

The Boreal Dilution Model (BDM) for separating natural from anthropogenic acidification during hydrological events was recently presented by Bishop et al. (2000). The BDM is based on the concept that during episodes the natural, pre-industrial acid-base chemistry dynamics result primarily from dilution of the buffering capacity by low ionic strength snowmelt/precipitation and input of dissolved organic carbon (DOC) with its acid functional groups.

The BDM is driven by the observed ANC ($ANC_{(obs,t)}$ (Eq. (1)) and a dilution index ($DI_{(t)}$; Eq. (2)) at any time 't' during the flow event. In the BDM the sum of base cations ($BC = 2^*[Ca^{2+}] + 2^*[Mg^{2+}] + [Na^+] + [K^+]$) are used as DI which is a measure of the natural dilution of both BC and the anthropogenically significant anions of strong mineral acids ($ANSA = 2^*[SO_4^{2-}] + [NO_3^-]$). Using Eq. (3) the natural, pre-industrial ANC ($ANC_{(preind,t)}$) can be predicted. The difference between the predicted pre-industrial $ANC_{(preind,t)}$ (Eq. (3)) and the observed $ANC_{(obs,t)}$ (Eq. (1)) is interpreted as the human influence on the ANC ($\Delta ANC_{(poll,t)}$, Eq. (4))) during the episode.

$$ANC_{(obs,t)} = 2^*[Ca^{2+}]_{(t)} + 2^*[Mg^{2+}]_{(t)} + [Na^{2+}]_{(t)} + [K^+]_{(t)} - [Cl^-]_{(t)} - 2^*[SO_4^{2-}]_{(t)} - [NO_3^-]_{(t)} = BC_{(t)} - ANSA_{(t)} - [Cl^-]_{(t)} \quad (1)$$

$$DI_{(t)} = BC_{(t)}/BC_{(base)} \quad (2)$$

$$ANC_{(preind,t)} = DI_{(t)} \times (BC_{(base)} - ANSA_{(base)}) - Cl^-_{(t)} \quad (3)$$

$$\Delta ANC_{(poll,t)} = ANC_{(preind,t)} - ANC_{(obs,t)} \quad (4)$$

The subscript 'base' denotes the baseflow condition preceding the high flow event.

4. The one-point model

Bishop et al. (2000) and Laudon and Hemond (2002) showed that there was a consistent relationship between the SO_4^{2-} winter deposition concentration and the average $\Delta ANC_{(poll,t)}$ of spring melt episodes from acid sensitive streams in northern Sweden. A similar correlation has been used in this paper using a linear regression (r^2 0.60, $n = 34$) between the average of the three consecutive samples that were most acid and the SO_4^{2-} winter deposition concentration for streams with baseflow ANC below 250 $\mu eq\ l^{-1}$ (Fig. 2). The most acid condition as well

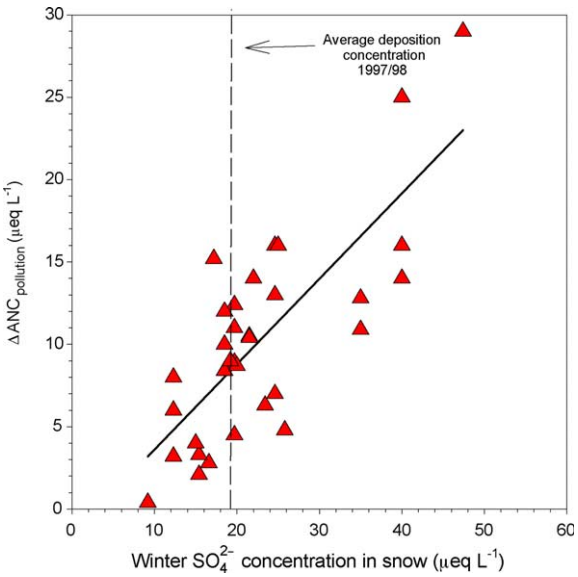

Fig. 2. Correlation between $\Delta ANC_{(poll,\ peak)}$ and SO_4^{2-} concentration in snow. Average deposition for the winter of 1997/1998 is shown.

as the largest ΔANC_{poll} have generally been found to coincide with peak spring melt runoff (Laudon, 2000).

The consistent correlation between winter S deposition concentration and the ΔANC_{poll} enables a prediction of the $\Delta ANC_{(poll,\ peak)}$ (Eq. 5) under the most acid condition during the spring runoff using the winter precipitation concentration of SO_4^{2-}, thus obviating the need for a time series of chemistry.

$$\Delta ANC_{(poll,peak)} = SO_{4\ winter}^{2-} \times 0.52 - 1.8 \quad (5)$$

To assess the severity of the anthropogenic component during spring melt runoff events, not only the $\Delta ANC_{(poll,\ peak)}$ but also the pre-industrial ANC at peak flow ($ANC_{(preind,\ peak)}$; Eq. (6)) and the predicted peakflow ANC ($ANC_{(pred,\ peak)}$; Eq. (7)) must be quantified (cf. 'Evaluation of Episodic Acidification' below). The quantification of $ANC_{(preind,\ peak)}$ in the pBDM (Eq. (6)) is almost analogous to that in the BDM (Eq. (3)). In the pBDM, the DI is an average BC dilution at peak flow associated with 53 spring melt runoff events from northern Sweden used in this study. Since DI was found to be significantly larger for the mountain sites (DI = 66%, std. dev. 11%; $n = 20$) than for the coastal and inland sites (DI = 53%, std. dev. 10%; $n = 33$), these two different DI:s were used in the Mountain and Coastal/Inland subregions. Another difference between the BDM and the pBDM is that the natural dynamics of Cl are accounted for in the BDM (Eq. (3)) but are not accounted for in the pBDM (Eq. (6)). It is not likely, though, that this is a serious flaw in the model since the introduced error would affect both $ANC_{(pred,\ peak)}$ and $ANC_{(preind,\ peak)}$ similarly; see section 'Evaluation of Episodic Acidification' below).

$$ANC_{(preind,peak)} = ANC_{(base)} \times DI_{(peak;average)} \quad (6)$$

$$ANC_{(pred,peak)} = ANC_{(preind,peak)} - \Delta ANC_{(poll,peak)} \quad (7)$$

The uncertainty introduced by transforming the full BDM to the baseflow based pBDM were tested using a Monte Carlo simulation in which two sources of uncertainty were tested simultaneously. The first uncertainty was in the linear correlation between the SO_4^{2-} deposition and the ΔANC_{poll} where the standard deviation in the ΔANC_{poll} residual was used to create a normally distributed population of correlation offsets for Eq. (5). The second uncertainty tested was that introduced by using a regional average DI. The larger standard deviation of the DI from the mountain sub-region was used to create a normally distributed population of DI values for Eq. (6). In the Monte Carlo simulation 10,000 random samples from the two parameter distributions were used. Other major assumptions evaluated in the use of the pBDM are that (1) BC is a reliable tool for simulating natural dilution (DI) during spring melt events, (2) winter lake ANC has not changed due to anthropogenic deposition (or the difference can be corrected for), and (3) lake ANC sampled during winter conditions can simulate the baseflow ANC (or the difference can be corrected for).

To test the assumption that the winter lake ANC has not been affected by anthropogenic deposition the Steady State Water Chemistry Model (SSWC) as implemented by Wilander et al. (1998) has been used together with background SO_4^{2-} estimates for the 1240 lakes. The SSWC is based on the concept of an 'F-factor' that represents the degree to which acid deposition is neutralized by the soil. Another important feature of the model is an estimate of the background SO_4^{2-} in surface water. A number of empirical approaches have been suggested for estimating the background SO_4^{2-} of surface waters in Scandinavia that correlate the sum of Ca and Mg to the natural background SO_4^{2-} (Wilander, 1994;

Andersson et al., 1999). Here we use the correlation by Andersson et al. (1999) since they specifically looked at the natural background SO_4^{2-} in northern Sweden using the same data set employed in this study.

The assumption that the winter lake ANC is an estimate of the winter baseflow ANC of streams has been tested by comparing volume weighted annual mean runoff ANC from five streams in northern Sweden (which is assumed to be the winter ANC in a downstream lake) and their baseflow ANC (H. Laudon, unpublished data). These data suggest that the winter baseflow ANC is approximately 20–30% higher then the annual average runoff ANC. An ANC correction of 25% has therefore been used to test the sensitivity of using the lake ANC to define the baseflow ANC.

5. Evaluation of the Episodic acidification

An acidification index (AI) to quantify the acidification status of Swedish surface waters based on the ANC concept was developed for the Swedish EPA (Wilander, 1999). The AI was initially developed as a guideline for determining liming candidates, but is here used to assess the present anthropogenic acidification component associated with spring flood episodes in northern Sweden. The AI is calculated as the ratio between $ANC_{(pred,\ peak)}$ and $ANC_{(preind,\ peak)}$ in individual surface waters (Eq. (8)).

$$AI = ANC_{(pred,peak)}/ANC_{(preind,peak)} \quad (8)$$

The Swedish EPA has separated AI into five classes according to the degree of acidification/sensitivity (Table 3) based on a large national survey of 4000 lakes

Table 3
Acidification index

Acidification class	$ANC_{(pred\text{-}obs,\ peak)}$/ $ANC_{(preind,\ peak)}$	Acidification index
1	1–0.75	No significant impact
2	0.75–0.50	Moderate impact
3	0.50–0.25	Large impact
4	0.25–0.10	Very large impact
5	<0.10	Extreme impact

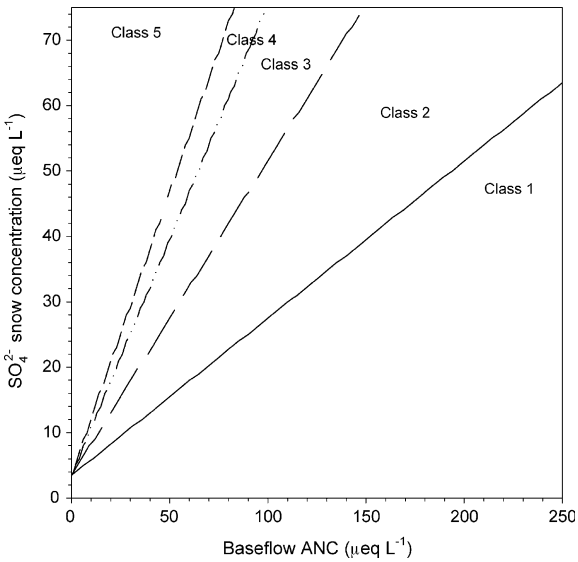

Fig. 3. The dependence of the Acidification Index (AI) on baseflow ANC and snow SO_4^{2-} concentration using the DI for the coastal and inland subregions.

and 700 streams in Sweden (Wilander et al. 1998). Because the AI depends on the ratio of predicted ANC and pre-industrial ANC during peak discharge, this ratio is controlled by the winter SO_4^{2-} concentration in snow and baseflow ANC (Fig. 3).

6. Regional predictions

The application of the pBDM to 1240 lakes from northern Sweden shows a generally low, but spatially variable response to the winter deposition 1997/1998. In northern Sweden as a whole, 6% of the lake population is subject to a degree of acidification that is classed between 'large' and 'extreme' during spring flood (Fig. 4; Table 4).

Division of the northern Sweden region into subregions reveals that the response to anthropogenic acid deposition associated with spring flood episodes was largest in the mountain areas despite the lowest anthropogenic deposition there. For the northern and southern Mountain sub-regions 9–15% of the area was classified as largely to extremely affected, while the figure for the rest of northern Sweden was between 2 and 4% (Table 5).

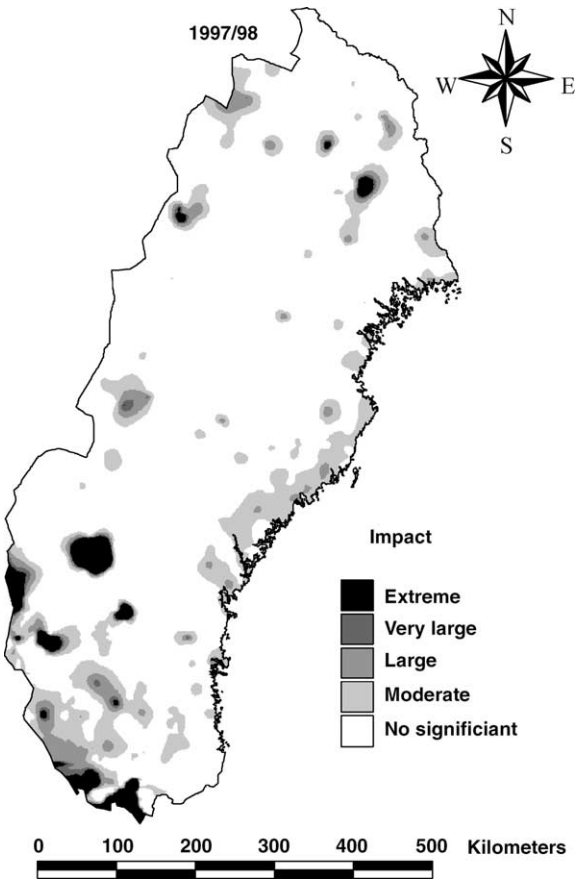

Fig. 4. Areas affected by different levels of anthropogenic episodic acidification during the 1998 spring flood.

Table 4
Model results and sensitivity of the *F*-factor and dilution correction

Acidification class	pBDM (%)	SSWC correction pBDM (%)	Lake dilution correction pBDM (%)
1	85 (11)	76 (10)	89 (11)
2	9 (7)	15 (8)	7 (6)
3	3 (2)	4 (3)	2 (1)
4	1 (1)	2 (1)	1 (1)
5	2 (2)	3 (2)	1 (1)

The number in parenthesis is the standard deviation derived from 10,000 Monte Carlo simulations randomizing a normal distribution of the uncertainty in dilution (± 0.11) and SO4 peak/ANC_{poll} correlation offset.

Table 5
Model results in sub-regions

Region	Acidification class	pBDM (%)	SSWC correction pBDM (%)	Lake dilution correction pBDM (%)
Southern Coast	1–2	95	92	96
	3–5	5	8	4
Southern Inland	1–2	94	89	94
	3–5	6	11	6
Southern Mountains	1–2	85	82	86
	3–5	15	18	14
Northern Coast	1–2	98	94	99
	3–5	2	6	1
Northern Inland	1–2	97	94	98
	3–5	3	6	2
Northern Mountains	1–2	91	89	94
	3–5	9	11	6

The Monte Carlo estimate of uncertainty in the pBDM is expressed as the standard deviation of lakes of the total population (Table 4). For all of Sweden, the average standard deviation in the number of lakes falling into class 1 was 11% while it was considerably less in the other classes (Table 4). A correction of the winter lake ANC for anthropogenic deposition (using the SSWC model) suggests that the assumption of an unaffected ANC could lead to under-predicting the number of lakes in the most affected classes (class 3–5) by less than 4 percentage points for the individual sub-regions (Table 5) and approximately 3 percentage points on average for all of northern Sweden (Table 4). The potential uncertainty introduced by the assumption that the winter lake ANC is equivalent to the winter baseflow ANC could lead to an over-prediction of the number of lakes in the most affected classes by less than 2 percentage points for the entire lake population (Table 4) and in general by less than 3 percentage points for the individual sub-regions (Table 5).

7. Discussion

The regional prediction of the anthropogenic deposition component driving the ANC decline associated with spring runoff events in northern Sweden demonstrates that the anthropogenic influence on the 1998 spring flood acidity was rather limited in most areas of northern Sweden on the regional scale of this study. Since the 1997/1998 winter deposition over northern Sweden is relatively typical for the latter half of the 1990's despite some interannual variations (Kindbom et al., 2000) these results are representative of the situation in the region during this period. However a general decline in acid deposition during the last two decades has resulted in measurable improvements in water chemistry in many lakes in southern Scandinavia (Stoddard et al., 1999; Fölster and Wilander, 2002). The results from this study extend such findings to the northern part of the country. The major difference is that since the anthropogenic component of acidity during spring flood is only a fraction of what it was a few decades ago, the recovery has been much faster and more complete (Laudon and Bishop, 2002a).

In the paper by Bishop et al. (2000) three major assumptions for BDM were presented, two of which are also of concern for the pBDM. The first of these assumptions is that BC is a reliable tool for simulating natural dilution during spring melt events. A critical review of this assumption is provided by Bishop et al (2000) and Laudon et al. (2001). Base cations have also been used in several other studies to quantify natural dilution of ANC during episodes (Molot et al., 1989; Kahl et al., 1992; DeWalle and Swistock et al. 1994). Although these references do not provide evidence that BC is a decisive measure of separating natural and anthropogenic acidity, the average DI of 53% for the coastal and inland regions (and the somewhat larger DI of 66% for the mountain regions) found in this study is consistent with the literature value of 50% based on hydrograph separation tracers used by Eshleman et al. (1995). A recent detailed hydrograph separation study in northern Sweden also confirms such approximation since BC and oxygen-18 gave almost identical hydrograph separations (Cory, 1999).

The second major assumption is that the baseflow ANC (or in this case the winter lake ANC) has not changed due to anthropogenic deposition. The validity of this assumption has been extensively tested and confirmed using paleolimnological reconstructions (Korsman, 1993; Korsman, 1999; Ek, 2000) soil studies (Tamm and Hallbäcken, 1988; Jacks, 1991; Eriksson et al., 1992) and Magic simulations (Kram et al., 2001).

Although the studies cited above argue against the need for a baseflow correction (except perhaps for the southern coastal sub-region (Stegman, 1990)), the Steady State Water Chemistry Model (SSWC) was used to estimate the sensitivity of this assumption. The result from the SSWC adjustment supports the assumption (and the paleolimnological findings) that only a marginal, if any, lake acidification of the winter lake ANC have occurred on a regional scale (Tables 4 and 5).

In the pBDM the winter baseflow ANC provides the benchmark from which the natural and anthropogenic ANC decline are predicted. A major assumption using lake water chemistry is therefore that the late winter lake ANC is analogous to the winter baseflow conditions of the streams with which the pBDM was calibrated. Even if the assumption that lake ANC has not changed due to anthropogenic acidification is correct, the residence time of water in lakes can be long enough to make lake winter chemistry more dilute than stream baseflow chemistry since the lake is influenced by dilution from antecedent precipitation/runoff events. If the more dilute lake winter chemistry is used as a surrogate for stream baseflow (as is the case in this application of the pBDM), then the anthropogenic influence on spring flood would be slightly overestimated. The fact that the two assumptions concerning the lake ANC (acidification and dilution) go in different directions strengthens the reliability of the results from this study.

While the BDM can be used to separate and quantify the natural and anthropogenic components of episodic acidification during hydrological episodes during any time of the year, provided that applicable data is available, the application of pBDM can only be expected to give a reasonable prediction with spring runoff events. A more mechanistic approach, such as the models proposed by Gerritsen et al. (1996) and Davies et al. (1999) which incorporate changes in discharge and antecedent precipitation is probably necessary for a regional prediction of rain driven

events due to a less proportional response between precipitation SO_4^{2-} and $\Delta ANC_{(poll,\ peak)}$ (Laudon and Bishop, 2002b). The reason that a predictive model such as the one used here or by Eshleman et al. (1995) works well with spring flood events and not with rain driven events is probably that the antecedent spring runoff conditions are influenced by less inter-annual variation compared to rain driven events where, for instance, dry periods can oxidize sulfur, rendering it more available for washout in a subsequent runoff event. Furthermore while low temperatures and soil frost will probably not prevent water from infiltrating the soil (Nyberg et al., 2001), the low temperature will prevent the soil from the same rate of chemical interaction as during summer and fall due to less oxidation in the time before the runoff event (as opposed to high temperature at the time of rain water infiltration). In the study region the lake ice cover usually persists from several months in the southern region to over six months in the northern region.

Although based on a large data set from surface waters in Sweden, the AI used in this paper only provides a qualitative measure of the acidification status. The advantage of using the AI to distinguish the degree of acidification of individual sites lies in the operational applicability of the index allowing for a consistent classification of the anthropogenic influence. Another advantage with AI, which improves its value, is that the sensitivity of streams increase with the decreasing baseflow ANC. This is appropriate since surface waters with the most acid baseflow conditions (lowest ANC) are the systems most predisposed to undergo biological damage as a result of anthropogenic acidification of spring runoff (Laudon, 2000).

In the calibration of the pBDM, only the acid sensitive streams (with a baseflow ANC below 250 $\mu eq\ l^{-1}$) were used to predict the anthropogenic component of ANC decline. If the more well buffered sites were incorporated in the correlation, a linear model, similar to Fig. 2, was also arrived at, but with less predictive power ($r^2 = 0.28, n = 53$). An important reason for the loss in correlation of snow SO_4^{2-} and $\Delta ANC_{(poll,\ peak)}$ in well buffered sites can be that high surface water ANC often coincides with natural bedrock/sediment sulfide bearing minerals. Since it is not possible to separate the release of natural SO_4^{2-} from atmospheric deposition of anthropogenic SO_4^{2-} using the BDM, a cutoff for the linear regression model at a baseflow ANC of 250 $\mu eq\ l^{-1}$ was used to define the upper limit of acid sensitive surface waters. As it is not likely that the more well buffered streams will respond differently to snow SO_4^{2-} (at least not with a significantly greater $\Delta ANC_{(poll,\ peak)}$ response) compared to the acid sensitive waters, the same linear model was used for these lakes (approximately 25% of the lakes had an ANC above 250 $\mu eq\ l^{-1}$). In order for a lake with an ANC above 250 $\mu eq\ l^{-1}$ to fall into an acidification class other than the least affected class 1, a SO_4^{2-} snow concentration of more than 65 $\mu eq\ l^{-1}$ is necessary (Fig. 3). No lakes experienced those deposition criteria during the winter of 1997/1998. A significantly different response from well-buffered lakes are thus not likely to alter the results from this study.

In the pBDM no account is taken of the deposition of NO_3^-. Although the nitrate has been found to be a significant driving mechanism of episodic acidification during spring runoff in northeastern USA (Galloway et al., 1987; Schaefer and Driscoll, 1993) results from the episodic acidification project in northern Sweden show no ANC decline due to nitrate during spring melt runoff events (Laudon et al. 2000).

The declining spatial extent of a large anthropogenic influence on spring flood acidity does not mean that spring ANC/pH decline is disappearing from northern Sweden. A large spring flood ANC decline (equivalent to the DI of ca. 50%) is a natural feature of aquatic ecosystems in this region. The relative increase in organic acids during spring flood also contributes to a natural pH decline (Ivarsson and Jansson, 1995; Laudon et al., 2001).

Since this empirical approach is based on the SO_4^{2-} winter deposition together with an approximation of the DI, the model can easily be applied to predict the anthropogenic component of ANC decline from winter baseflow chemistry. The approach taken in this paper is similar to the 'index approach' presented by Eshleman (1988) and later applied in a regional scale study (Eshleman et al., 1995). While the main object in the study by Eshleman et al. (1995) was to predict the minimum ANC during snow melt runoff events, the purpose of pBDM is to predict the anthropogenic component of the ANC decline associated with the spring runoff. Although

the difference between the models might appear marginal, the separation of the anthropogenic contribution from the natural ANC decline and not only minimum ANC is of importance when assessing the human influence on the surface water chemistry in different regions. A quantification of the anthropogenic component is especially important when remediation such as liming is being planed in order to avoid destruction of naturally acid ecosystems.

There is an inherent loss of accuracy when predicting the chemistry of individual streams when moving from the scale of a well studied catchment to a regional prediction. The sacrifice in predictability is, however, compensated for by the more synoptic overview gained on the regional scale. While the empirical model approach taken in this paper is powerful in that it can be applied to regional data sets, the analogous approach presented by Eshleman (1988) has been subject to much interest and debate (Schaefer et al., 1990; Schaefer and Driscoll, 1992; Eshleman, 1992; Eshleman et al., 1995). A concern raised by Schaefer et al. (1990); Schaefer and Driscoll (1992), to the empirical index approach suggested by Eshleman (1988), is that hydrological flow pathways are a critical and highly variable factor which needs to be dealt with using process-oriented models for a proper assessment of episodic acidification. However, since only the largest and most detailed data sets are sufficient to calibrate the even simplest process-oriented hydrochemical models, empirical approaches are necessary in order to provide a regional prediction of the effect of acidification during snow melt episodes. Since the empiricisms in the pBDM are derived from detailed hydrochemical time series, it has been possible to assess the uncertainties introduced by generalizing the hydrological response on the anthropogenic component of spring ANC decline. This should increase the reliability of the model results, and point the way for using detailed studies to support more empirical operational models.

8. Implications of model results

The results of this study show that the present human impact on spring flood hydrochemistry in northern Sweden is generally limited. This has important implications for planning national liming strategies in the region. The results also have important international implications for future reduction of air pollutants over Europe. The decline in SO_4^{2-} over northern Sweden so far has probably led to a reduction in the acidification pressure over northern Sweden during the last two decades. A further reduction in acid deposition, however, is needed before all surface waters in the region are free from acid episodes driven by anthropogenic acidification.

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