ESTIMATION OF BACKGROUND SULPHATE CONCENTRATIONS IN NATURAL SURFACE WATERS IN SWEDEN

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Abstract. In order to measure the extent of acidification the background, 'preindustrial' conditions must be known. An equation for the estimation of background concentrations of sulphate in surface water in Norway was proposed by Henriksen. When applied on data from the Swedish lake survey in 1990 it was found that the calculated background concentrations exceeded those measured for about one-third of the lakes. The proposed revision is based on a background concentration in precipitation and an estimated contribution from weathering, the latter associated with base cations. Three different approaches were tested to establish the contribution from weathering; geochemical ratios or groundwater chemistry data as a basis, and historical data on denudification. The weathering calculated from groundwater chemistry data seems to give the best estimate of the background sulphate concentration in surface water. Organic matter as source or sink of sulphur is discussed and considered negligible.

1. Introduction

Background concentrations of different constituents in lake water are important to establish in order to be able to evaluate the anthropogenic influence. Meybeck and Helmer (1989) state that the determination of natural background concentrations and loads of pre-industrial times is a major challenge remaining in river geochemistry.

Background concentrations of sulphate are of special importance for the evaluation of acidification of natural water as made by Henriksen (1984) for preacidification alkalinity. Later the way of calculation was further developed and applied for the estimation of critical load values for acid deposition on aquatic ecosystems with a steady state water chemistry model (Brakke *et al.*, 1990; ECE, 1990; Henriksen *et al.*, 1990) which requires as one input a value of the background sulphate concentration. This is one of the uncertainties when applying the model (Husar *et al.*, 1990), which thus requires further studies. In the case of acidic water, sources are generally considered namely sulphate and organic acids (Kortelainen and Mannio, 1990; Dupont, 1992). As the importance of these sources is difficult to evaluate, an estimate of the background sulphate concentration might contribute to a better understanding of the significance of the sources of acid.

Two sources of sulphate are recognized, namely deposition and weathering. In the following an attempt to calculate background concentrations in natural water will be made. Starting from an equation proposed by Henriksen (ECE, 1990), which is based on the concentration of base cations in the water, different alternatives,

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described below, have been tried in order to modify the equation for the calculation of background sulphate concentrations in Swedish lake water.

2. Materials and Methods

2.1. SWEDISH LAKE SURVEY

During the winter-early spring of 1990 a lake survey covering 4018 lakes was made in Sweden, comprising about 5% of Swedish lakes. The lakes were chosen on a statistical basis in five strata according to lake size. Most of the about 380 lakes belonging to the two largest size classes (10 to 100 and >100 km^2) were sampled. For the other three strata, 40 lakes per county were selected at random (Bernes, 1991). The situation of the lakes are shown in Figure 1.

Liming activities have directly or indirectly affected some lakes which were excluded, leaving 3171 lakes for the evaluation. Sampling was usually performed in the middle of the ice-covered lake at a water depth of 2 m. Samples were immediately sent to the laboratory in Uppsala for analysis. On all samples, pH, conductivity, cations, anions and absorbance at 420 nm(color) were determined using ICP OES for the cations, titration for alkalinity (ISO) and ion chromatography (with suppressor) for chloride and sulphate. Silica was analyzed on one forth of the samples by the molybdate method. Analytical quality control was applied throughout the measurement period.

Non-marine concentrations of base cations BC*, and sulphate were calculated according to ratios in seawater, based on chloride concentrations (c.f. ECE, 1990)

$$[BC^*] = [BC] - 0.235[Cl] \; [SO_4^*] = [SO_4] - 0.103[Cl]$$

2.2. ADDITIONAL WATER CHEMISTRY DATA

Groundwater chemistry data were selected from the Swedish groundwater network (SGU, 1985). Results from 20 areas were used, 76% of the total of 138 samples, collected between 1982 and 1990, came from four stations in the northern Sweden (counties 24 and 25). All these aquifers were situated in moraine or gravel. The analyses of interest were made with ion chromatography and emission spectrophotometer.

A survey of 52 lakes in the Abisko area in northernmost Sweden (county 25) was made in August 1981. The samples were analyzed at the laboratory using atomic absorption for the cations. For sulphate the Mackereth method was used, which leads to some overestimation when organic anions are present (Gorham and Detenbeck, 1986).

Older data have been collected from three references. Lohammar (1938) analyzed samples concentrated by evaporation for the determination of cations by emission spectrography and sulphate by gravimetric determination of of barium sulphate. The method was checked by Andersson (1980) and found to have a good precision and accuracy. The methods used by J. V. Eriksson (1929) and Sondén

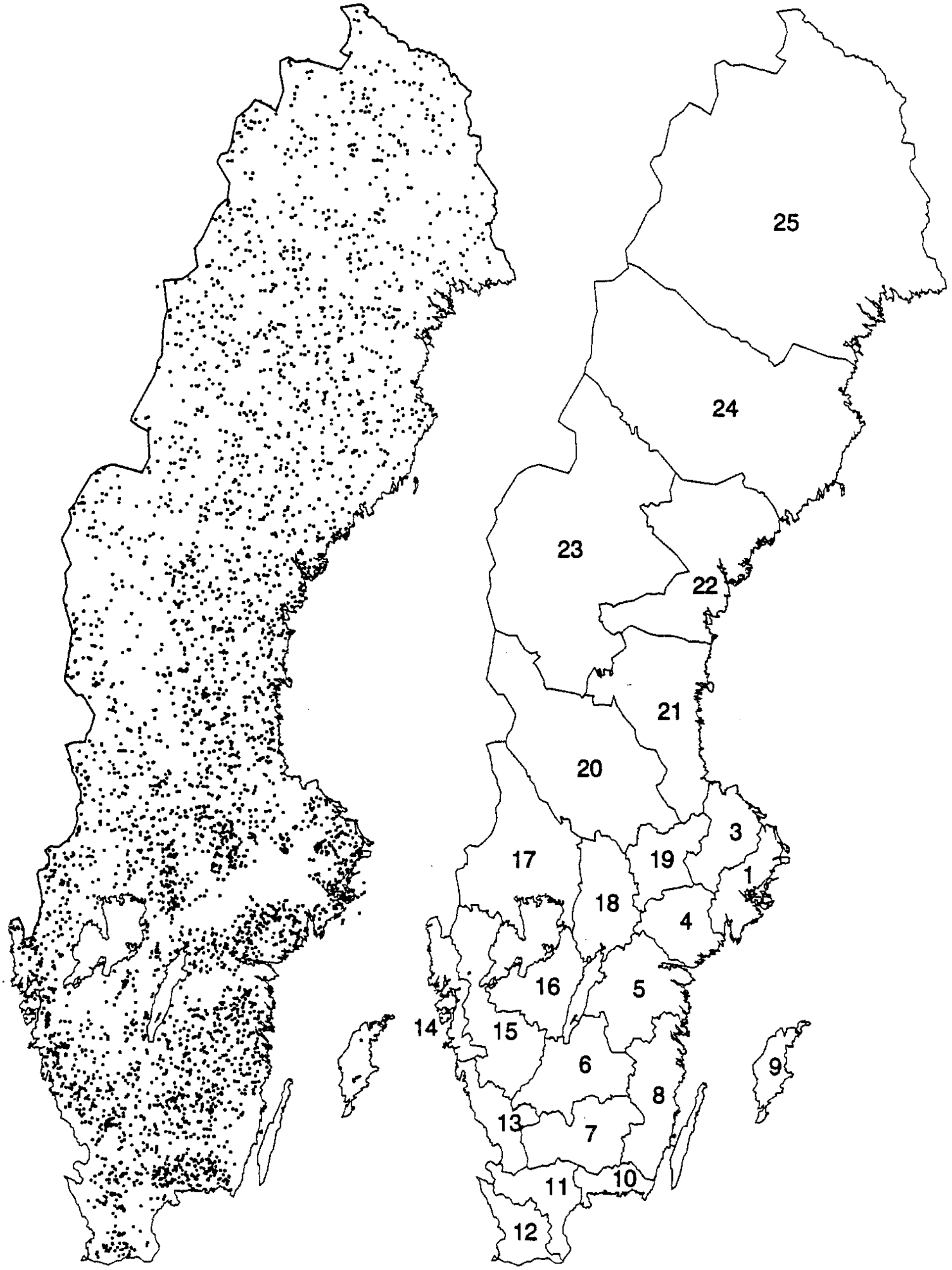

Fig. 1. Map over location of Swedish lakes sampled and counties.

(1914) are not well described. Attempts have been made to quality check the data by ion balance and comparison with dissolved solids and erroneous data have been excluded.

3. Application of the Henriksen Equation

One attempt to estimate the background sulphate concentration was made by Brakke *et al.* (1989). Later, Henriksen (ECE, 1990) published an equation for the calculation based on data from relatively unaffected Norwegian lakes (in μeq L^{-1}).

$$[SO_4^*{}_o] = 15 + 0.16[BC^*] \qquad n = 148 \qquad r = 0.38$$

where $[SO_4^*{}_o]$ = non-marine background sulphate concentration and $[BC^*]$ = non-marine base cation (Ca^* + Mg^*) concentration. The author thus considered two sources, one airborne (unaccompanied by base cations) with a fixed concentration and one from weathering (related to the base cation concentration). Henriksen considered the equation to be valid for Norway, and stressed that it should be applied to other areas with caution. Furthermore, he pointed out that the relation would be improved if the data set could be split up into different mineralogical groups. This has not been possible to do with the Swedish data set. Recently Posch *et al.* (1993) published a similar equation based on Finnish data;

$$[SO_4^*{}_o] = 14 + 0.10[BC^*] \qquad n = 61 \qquad r^2 = 0.29.$$

About 30% of the Swedish lakes were found to have a Henriksen background concentration higher than that observed for the lake (Figure 2). This discrepancy led to the search for an alternative equation to calculate the background concentration of sulphate in Swedish lakes.

4. Alternative ways to estimate the background concentration

As Henriksen (ECE, 1990) stated, two sources for the sulphate can be recognized, namely the atmosphere (precipitation) and pedosphere (weathering). In the following the two sources will be discussed in detail.

4.1. SULPHATE INPUTS FROM PRECIPITATION

Most input-output studies made in Norway and Sweden have shown that little or no sulphate is retained or released in the area (E. Eriksson, 1988; cf. Mast and Drewer, 1990). This situation is likely to be the case under-industrial conditions, when adsorption processes were in equilibrium with deposition and when the soil/rock had a low concentration of S.

The contribution from precipitation requires a measured or estimated value of the background concentration of sulphate in the precipitation and an estimate of the concentrating caused by evapotranspiration. In Sweden the measurements of principitation chemistry started in November 1984 (Egnér and Eriksson, 1955). However the earlier determinations of sulphate are considered to be erroneous. Rodhe (1972) estimated the background sulphate deposition in northern Scandinavia to be about 16 μeq m^{-2}, y^{-1}, equal to a concentration in precipitation of

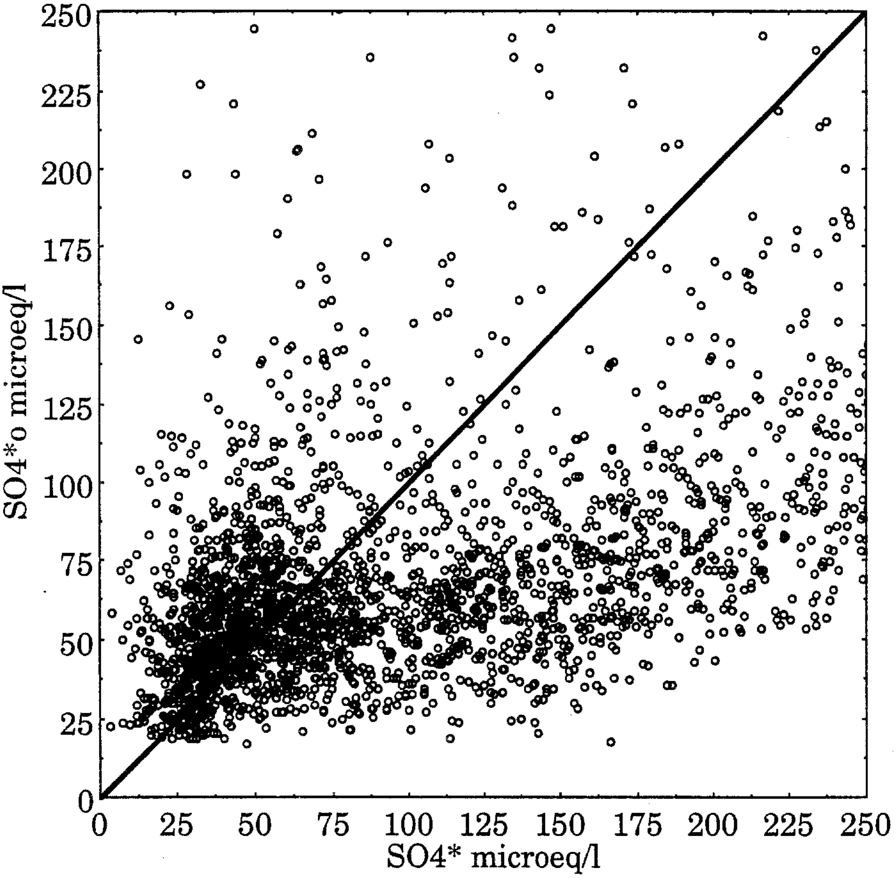

Fig. 2. A comparison between observed sulphate concentrations in Swedish lake water and background concentration calculated according to Henriksen (Equation (1)). The line gives the 1:1 relation.

21 μeq L^{-1} (based on 745 mm annual precipitation) and corresponding to a lake water concentration of 31 μeq L^{-1} (based on a 500 mm runoff). These relatively high results indicated a need for further study.

Precipitation chemistry in remote areas, little affected by air pollution, could be a basis for the estimation of background sulphate concentrations. For the purpose of this study it is reasonable to assume that pristine concentrations would be universal.

A set of data on sulphate concentrations in precipitation is presented in Table I. The precipitation at these more or less remote areas is still likely to be affected by masses with trajectories from distant sources of pollution and therefore the 'present remote area value' should be based on the lower measurements cited. The median value for the data presented is about 5 μeq L^{-1}.

Even for remote and locally unpolluted areas there is reason to consider the global spread of pollutants. Hallberg (1976) calculated the present terrestrial deposition to 60 Tg yr^{-1} as compared to a prehistorical 21, thus a threefold increase. A

TABLE I

Concentrations of SO_4 (μeq L^{-1}) in precipitation at remote sites

Area	Concentration	Reference
East Greenland	9.3 (21)	Keiding and Heidam, 1986
Alaska	7.2-12.1	Keiding and Heidam, 1986
Alaska, Point Barrow	4.9	Keiding and Heidam, 1986
Remote areas	3–10	Galloway *et al.*, 1984
Remote marine areas	4.1	Galloway *et al.*, 1989
North Atlantic	4.1	Buijsman *et al.*, 1991
W. Canada, snow core	11 (4–35)	Shewchuck, 1986
W. Canada, event rain	13 (10–18)	Shewchuck, 1986.
W. Canada	12–16	Barrie and Hales, 1984
N. Finland, snowpack	12	Soveri, 1985
USA, Sierra Nevada	4.3	Williams and Melack, 1991
Greenland ice		
Preindustrial	1.0	Dayan *et al.*, 1985
Recent	2.5	Dayan *et al.*, 1985
Preindindustrial	0.8±0.1	Laj *et al.*, 1992
Recent	2.0±0.4	Laj *et al.*, 1992

calculation of the biogenic and anthropogenic global S emission by Möller (1984) suggested that the present emission is about twice the biogenic (70 Tg yr^{-1}).

Already at the beginning of this century the anthropogenic emission in Euroope was about 10 Tg yr^{-1} (Möller *loc. cit.*) as compared to a global level of 14, a fact which underlines the extremely uneven geographical distribution both in emission and most likely deposition. Even through there is substantial difficulty in transferring emission data to deposition, the latter may be estimated to have increased by at least a factor of 2 in Sweden. This value equals that suggested by Galloway *et al.* (1984) for remote areas of north America.

If this 'pollution factor' is applied on the 'remote area sulphate concentration' of 5 μeq L^{-1} as given above, the estimate of the background non-marine sulphate concentration in precipitation will be about 2.5 μeq L^{-1}. Even such a low value is high in comparison with measurements of pre-industrial sections of the Greenland ice as shown in Table I (Dayan *et al.*, 1985; Lai *et al.*, 1992).

For Finland, Tuovinen *et al.* (1990) proposed that 'roughly 1/3 of the background used for sulphate in the EMEP S model [0.3 mg S/l] is of natural origin,' thus about 6 μeq L^{-1}. Lepistö *et al.* (1988) utilized emission data for the calculation of the background concentration in precipitation for Lake Yli-Knuutila, but in the case of Lake Liuhapuro the calculation was based on the present chloride deposition and the seasalt composition, thus considering seasalt as the only source of sulphate

TABLE II

Comparison between different calculations of background sulphate. Measured (1984) and calculated SO_4 (1844) concentrations in deposition at two lakes in Finland (Lepistö *et al.*, 1988) and in stream water (Kämäri *et al.*, 1990). Alternative equations are given in the text

Lake	SO_4 concentration μeq L^{-1}							
	Deposition		Stream water					
	1984	1844	1984	1844	Henriksen	Alt 1	Alt 2	Alt 3
Yli-Knuutila	61.1	1.8	585	13	13	8	39	83
Liuhapuro	53.3	0.9	82	2	33	6	11	16

(Table II). These two calculations of sulphate concentrations in the deposition result in only a few μeq L^{-1}, well in line with the estimate made above of about 2.5 μeq L^{-1}.

In order to transfer the concentration in precipitation to a potential surface water concentration, a correction has to be made for the evapotranspiration raising the concentration of sulphate as well as other ions. The concentrating effect due to this process varies throughout Sweden, with higher values in the southern (warmer) part of about 3 and the lowest in northern parts of round 1.5. B. Eriksson (1983) gave a mean evapotranspiration value of 43% of the precipitation for Sweden. Thus the concentration value in precipitation has to be multiplied by a factor of 1.75 (100/(100–43)), leading to a background preindustrial concentration in lakewater from precipitation of about 5 μeq L^{-1}.

4.2. SULPHATE INPUTS FROM WEATHERING

Weathering as a source of S could be estimated by the application of ratios for soil or rock between the concentration of S and one or more selected reference components, which are not, or only little, affected by anthropogenic activity. Alternatively the ratio may be based on the composition of water mainly affected by weathering. Such a calculation must be based on a relationship between the sulphate concentration and the concentration of a 'stable' component in the water such as base cations either as a sum BC (ECE, 1990) or individually, alternatively silica could be used (Odén and Ahl, 1978). However, seasonally the concentration of silica varies widely in surface water due to biological processes (*e.g.* Wetzel, 1975) and thus such a ratio can only be applied on winter data.

4.2.1. Geological ratios as a basis

The contribution of S from the geosphere to lake water through weathering may be calculated from the composition of the geosphere. This implies more or less fixed ratios between the concentration of S and that of the chosen component

TABLE III

Ratios between S and other elements (molar basis) (Krauskopf, 1979)

	Earth crust	Granite	Basalt	Shale
S/Si	0.00081	0.00073	0.00091	0.00081
S/Na	0.0108	0.0096	0.0132	0.0333
S/Ca	0.0063	0.0169	0.0037	0.0088
S/Mg	0.0113	0.0169	0.0056	0.0164
S/BC	0.0041	0.0181	0.0022	0.0058

TABLE IV

Calculated median concentrations of sulphate in lake water due to weathering based on geological ratios (equivalent) and median concentrations in Swedish lake water

Element	Facor	Median conc. μeq L^{-1}	Calculated concentration SO_4 μeq L^{-1}
Si	0.056[a]	1.8[b]	0.10
Na	0.020	99	2.0
Ca	0.006	234	1.9
Mg	0.012	90	1.1
BC	0.005	321	1.6

[a] μeq mg^{-1}.
[b] mg L^{-1}.

in weathering material. But it is worth noting that no S minerals contain silica and few of them contain any of the base cations. The implication of this is that in a strict sense a relationship based on mineral composition cannot be applied However in spite of this, a more general approach has to be tried. The crustal average concentration of S is 260 ppm and ultramafic, basalt and granite have concentrations varying from 250 to 300 ppm (Krauskopf, 1979). A set of ratios is presented in Table III.

The author is aware of only few Swedish data on the S concentration in rock. For the Gårdsjön area on the Swedish westcoast, Melkerud (1983) reported the mineral composition in granodiorite (with a S concentration of 0.02% or less) giving S/Si and S/BC ratios of 0.0006 ± 0.0001 and 0.0056 ± 0.0022, respectively.

The results obtained when using the lake survey data set give very low estimates of the sulphate concentration. This is described in Table IV where the 50th percentile lake concentration values for Si, Ca, Na, and Mg and a chosen set of ratios were used to estimate concentrations of S released from the soil/rock. The results are quite similar with the exception of that based on silica, which gives an extremely low value possibly due to incongruent weathering, leaving silica-rich minerals. In Swedish lake water, the median sulphate concentration is 132 μeq L^{-1}, thus being about 100 times higher than that calculated from Si weathering alone. For lakes less affected by acidic rain ($SO_4^* > 20$ μeq L^{-1}, n = 70), the calculation gives an estimated contribution through weathering of 0.017 μeq L^{-1}, which is about 60 times less than the actual concentration.

Based on the geological ratios we chose 0.005 as the equivalent ratio S/BC to be applied in a revised equation.

4.2.2. *Groundwater data as a basis for weathering estimation*

Groundwater chemistry should be a better indicator than the elementary composition of the weathering conditions of the soil and rock. Selected data from the Swedish groundwater network (SGU, 1985), mainly from the northern part of Sweden with comparatively low sulphate deposition, was used for this purpose. Only base cations were used as a basis for the calculation since no silica data were available, leading to the following equation.

$$SO_4^* = 0.053BC^* + 72 \qquad n = 138 \qquad r^2 = 0.18.$$

A detailed examination of data from separate stations showed a high slope value of 0.31 for Abisko with shale rock, while three other nearby stations with granite or other crystalline rock had slopes between 0.03 and 0.073. This underlines the sigificance of the different rock/soil types in governing the weathering. Using these data on groundwater composition, a general slope value of 0.05 for the contribution from soil/rock seems reasonable. This is about 10 times higher than that based on soil/rock composition (see 4.2.1), a situation likely to be caused by the uneven presence of sulphide minerals in rock and their properties to be easily weathered.

4.2.3. *Historical data as a basis for weathering estimation*

Data from periods less affected by acidic precipitation could contribute to the evaluation of background sulphate concentrations. Historical data for Swedish fresh waters are available from a few documents. The most well known is the study of Swedish rivers covering the period 1990–1925 and published by J. V. Eriksson (1929). For lakes, the best source of information is the study made by Lohammar (1938), who sampled lakes in different areas of Sweden and published results from fairly complete analyses. A third source of information is the publication by Sondén (1914), who presented data from varying analyses.

It is important that the data set used is based on aquatic systems which were not polluted at the time of study and may be vulnerable to acidification. As most of the acidified or potentially acidified lakes are situated above the highest marine limit,

TABLE V

'Historical' ratios between sulphate and base cation concentrations (equivalent basis) for natural surface water

Slope SO_4^*	Reference		
0.131±0.03	> 100 m asl, $BC^* < 0.5$ meq L^{-1}	0.20	Lohammar, 1938
0.099±0.016	> 100 m asl, $BC^* < 1$ meq L^{-1}	0.29	Lohammar, 1938
0.019±0.02(Ca^*)	lowland rivers excluded	0.015	J. V. Eriksson, 1929
0.16±0.10	lowland rivers excluded	0.14	Sondén, 1914

this was used as one restriction for the data set used. Furthermore, such lakes have low concentrations of base cations and this was the second restriction applied. The results are shown in Table V.

A reasonable estimate of the slope based on these calculations could be 0.10 SO_4^*.

4.3. THE FUNCTION OF ORGANIC MATTER

The calculations made above refer to water without organic matter and a substantial number of Swedish lakes have high concentrations of humic substances. The organic matter in soil could function as a sink for deposition S, while a release of organic material to water would then naturally be followed by S release as well.

Peat may contain relatively high concentrations of S, 1-2% in swampy areas, mainly in the form of sulphides (Troedsson and Nykvist, 1973) while a median S concentration of 0.2% (dry weight) was found for peat mined in Sweden (SCB, 1989). For Spagnum peat in southern Sweden, Assarsson (1961) found concentrations ranging from 0.06 to 0.17%. SCB (1989) estimated the growth of peat resources to be 63×10^6 t dryweight yr^{-1}. With a concentration of 0.2%, the average uptake in Sweden would be 0.1 g S m^{-2}, yr^{-1} ($\approx$ 20 meq m^{-2}, yr^{-1}). This is likely to be an overestimate as it is close to the present level of S deposition in many areas. In Finland, Huttunen and Karhu (1981) found a fixation in surface peat of 19 to 25% of the S deposited. It is clear that soil organic matter may function as a sink for deposited S.

A lowering of the groundwater level (such as by forest ditching) will reverse this process, allowing oxygen penetration into the peat and subsequent sulphide oxidation as well as releasing organic matter. This justifies the inclusion of organic matter as a source of S. The amount of S attributed to organic matter may be calculated from the concentration of organic C. Bringmark (pers. comm.) determined the concentrations of S and C in the mor layer in 17 forested areas of Sweden giving a mean SO_4/TOC ratio of 0.21 μeq mg^{-1}. The 50th percentile concentration of organic C in Swedish lake water is 7.5 mg L^{-1}, which would imply a sulphate leakage of 1.5 μeq L^{-1}, thus being less than 2% of the present median sulphate concentration. For 95% of the lakes, the estimated contribution to the sulphate concentration is less than 5% of the actual concentration. A detailed study of the situation in lakes in northern Sweden, with a reasonably low and uniform deposition, gives varying and weak indications for both sink and source processes. It is thus not possible to ascertain the role of organic matter in soil and water on the circulation of S, but it is likely to be small.

5. Discussion

Application of the equation prepared by Henriksen (ECE, 1990) on the Swedish data set resulted in about 30% of the lakes with background concentrations higher than that observed for the lake, specially in the lower range of concentration (Figure 2). This condition is even more pronounced in the northern parts of Sweden.

The literature review of background concentrations of sulphate deposition indicates that it is less than the intercept value of 15 μeq L^{-1} used by Henriksen for the non-marine background concentration in lake water and a value of 5 μeq L^{-1} has been chosen as the most likely alternative.

The weathering component (slope) was, depending of the basis for calculation, estimated to be 0.005, 0.05, and 0.10 respectively leading to three alternative equations for estimation of background sulphate concentrations with the exclusion of organic matter as a carrier of S:

$$Alt1 \qquad [SO_{4\ o}^{*}] = 5 + 0.005[BC^{*}]$$

$$Alt2 \qquad [SO_{4\ o}^{*}] = 5 + 0.05[BS^{*}]$$

$$Alt3 \qquad [SO_{4\ o}^{*}] = 5 + 0.10[BC^{*}]\,.$$

The results obtained with the three alternative equations are shown in Figures 3 to 5. All give substantially lower estimates of the background concentration ($SO_{4\ o}^{*}$) than the Henriksen equation. Alternative 1 gives very low estimates, while using Alternative 3 results in some estimates with concentrations higher than those observed. Since this Alternative is based on historical data, such a result could be caused by high anthropogenic emissions already at the beginning of the century (Möller, 1984), leading to elevated sulphate concentrations in the water and a slope value that is too high.

Table VI shows detailed results for some of the counties. The first two, Blekinge and Kronoberg (7 and 10), are heavily affected by acidic deposition, while the counties situated in the northern part of Sweden (23–25), have a low deposition. For the four northernmost counties were the $SO_{4\ o}^{*}$ estimates using the Henriksen equation higher than those measured (negative differences) in 70% of the lakes, while application of Alternatives 2 and 3 resulted on overestimates in 5 and 25% of the cases respectively. A comparison between the the sulphate concentrations in present deposition and the difference between present lake water concentration and

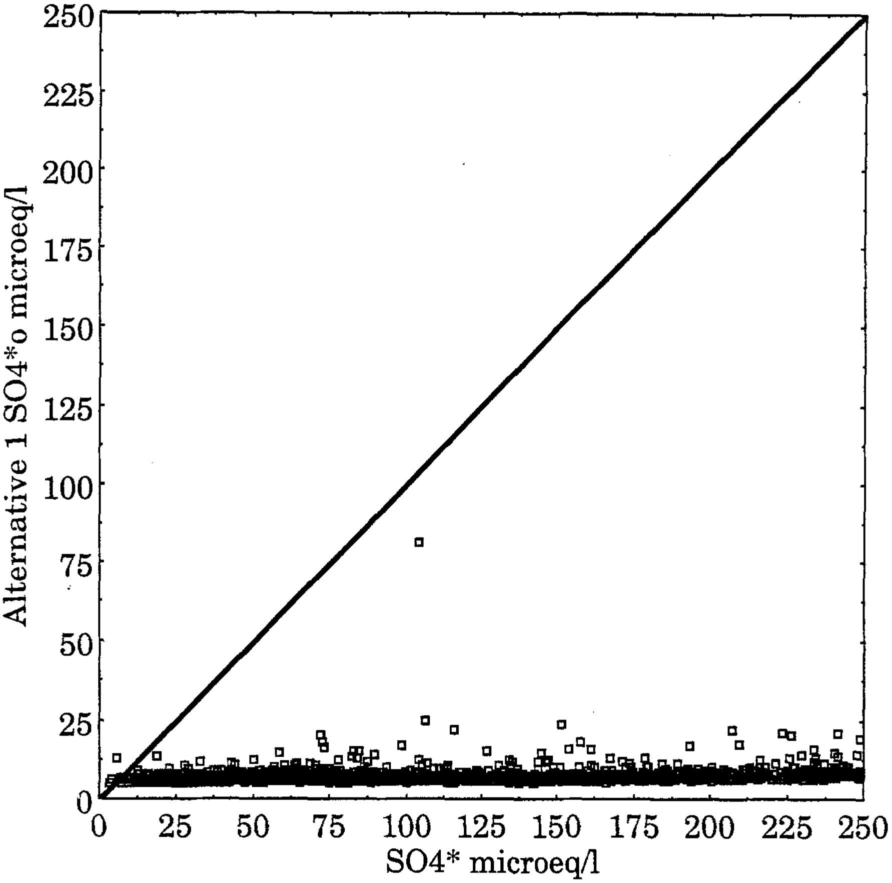

Fig. 3. A comparison between observed sulphate concentrations in Swedish lake water and background concentration calculated according to Alternative 1. The line gives the 1:1 relation.

background concentration could be used to validate the alternatives. For the most affected counties no conclusion can be drawn, while results for the less affected counties (23 to 25) support Alternatives 1 and 2.

Data from a separate survey of lakes in Abisko area (county 25) showed such a large variation that no indication about the validity of the alternatives can be found. As an example the background concentrations according to Henriksen varied between −112 and 169 μeq L^{-1}.

For a 'pristine' area in mid-Norway with a measured SO_4^* concentration in the precipitation of 8 μeq L^{-1} Christophersen *et al.* (1990) reported a measured stream water concentration of 14 μeq L^{-1}. Calculations using the equations of Henriksen and Alternatives 1 to 3 results in $SO_{4\,o}^*$ estimates of 15, 5, 6, and 7 μeq L^{-1}, respectively. Obviously all Alternatives give estimates in the range of a background concentration. Three independent estimates of the background concentration of sulphate in the Nordic countries are known to the author. Odén

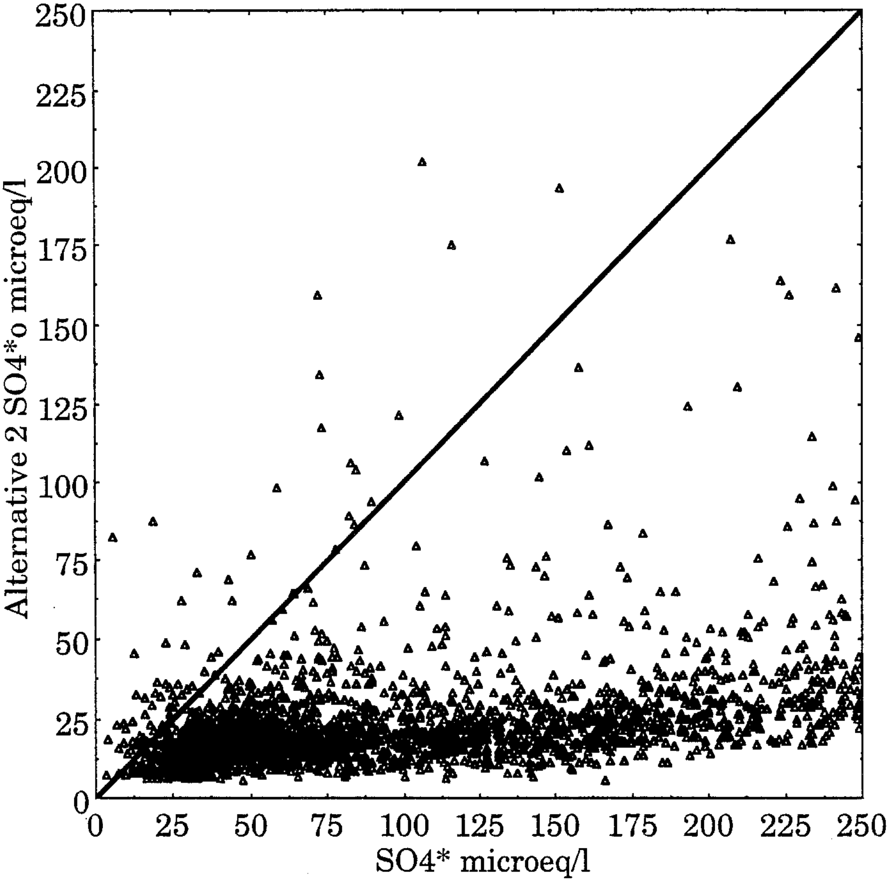

Fig. 4. A comparison between observed sulphate concentrations in Swedish lake water and background concentration calculated according to Alternative 2. The line gives the 1:1 relation.

and Ahl (1978) computed for Sweden the release of S due to weathering. They based their calculations on experimental leaching of 48 Swedish soil samples using 0.1 mol L^{-1} HCl and presented results relative to total mass of weathering products and silica leading to a mean quotient of 0.74 μeq SO_4/mg Si. This is about 10 times higher than that for the rock (see Table III), a situation expected and caused by inhomogeneous weathering rates of the soil minerals. A comparison of the outcome of this calculation with the quotients for weathering (slope values of Henriksen and Alternatives 1 to 3 equations) for the Swedish lake water leads to values lower by a factor of 64, 2, 20, and 40 times respectively. The differences may be caused by errors in the sulphate analysis (probably the thorin method, J. Bergholm pers. comm.) but Alternative 1 gives the closer result.

Wright *et al.* (1986) used OECD emission values as one input in the MAGIC model for the calculation of water chemistry in four lakes, of which two are in the Nordic area. The data given in Table VII clearly show the impact of anthropogenic

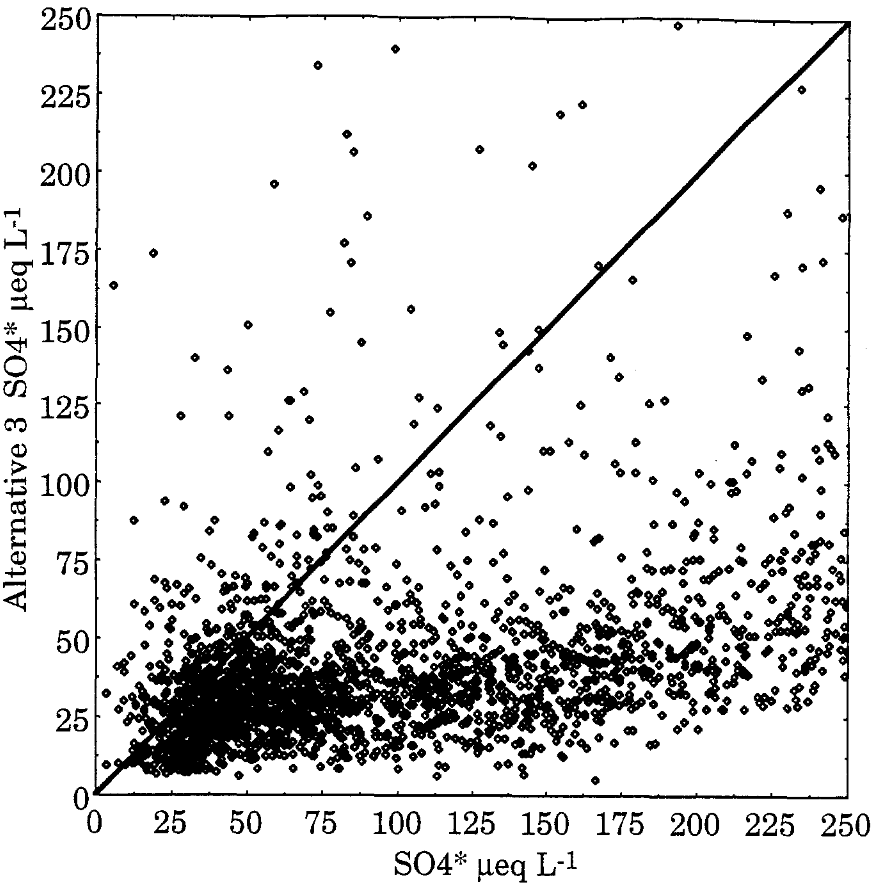

Fig. 5. A comparison between observed sulphate concentrations in Swedish lake water and background concentration calculated according to Alternative 3. The line gives the 1:1 relation.

sulphur sources on water chemistry already at the beginning of this century. For Lake Gårdsjön Alternative 3 gives a result most equal to that of the MAGIC model.

The third example deals with two lakes in Finland. Kämäri *et al.* (1990) applied the MAGIC model and calculated and applied values for sulphate concentrations in the deposition for the modeling of long-term acidification. Based on stream water BC* concentrations measured 1984, results from different estimates are given in Table II. Most similar to the MAGIC results (Kämäri *et al.*, 1990) are those of Alternative 1, while the others deviate substantially. Based on the above Alternative 2 may be considered as giving the best estimate of the background sulphate concentration ($SO_{4\,o}^{*}$). However there seems to be a regional variation with Alternative 1 as more adequate for the Northern, less acidified parts of Sweden and Alternative 2 for the Southern parts. Such a difference may be caused by an intensified weathering due to acid deposition (Henriksen, 1984; Brakke *et al.*, 1990; Sverdrup, 1990).

TABLE VI

Mean difference between measured sulphate concentrations (SO_4^*) and calculated background values ($SO_{4\,o}^*$) in swedish lake water. Present sulphate concentration in deposition according to Granat (1990)

	Difference $SO_{4\,o}^* - SO_{4\,o}^*$ μeq L^{-1} Calculation method					Present deposition SO_4^* μeq L^{-1}
County	Henriksen	Alt 1	Alt 2	Alt 3	n	
7	184	255	237	218	79	70–>80
10	220	310	278	253	61	>80
17	55	94	87	77	116	40–60
18	93	173	153	130	71	50–60
19	61	163	137	107	85	40–70
20	28	86	72	56	208	40–60
21	14	95	74	52	167	40–60
22	27	102	83	62	147	40–60
23	–18	35	21	7	283	20–30
25	-4	45	32	19	365	20–50
25	–10	32	23	12	487	20–40

TABLE VII

Measured (a) and calculated (b) SO_4^* concentrations in two lakes, Hovatn (Norway) and Gårdsjön (Sweden) (Wright *et al.*, 1986) and calculated according to alternatives given in the text

	SO_4^*						
Lake	1984 (a)	1904 (b)	1844 (b)	Henriksen	Alt 1	Alt 2	Alt 3
Hovvatn	90	35	2.5	–	–	–	–
Gårdsjön	180	80	21	36	6	11	18

6. Conclusion

As we have no values of true pre-industrial sulphate concentrations is there no fully objective way available for validation of different alternatives, but there is good reason to consider equations giving estimates higher than present concentrations as less adequate, which is often the case with the Henriksen equation (Equation 1, Figure 2), and less frequent with Alternative 3 (Figure 5). Alternative 1 often gives estimates lower than can be expected (Table VI), and thus Alternative 2 (Figure 4) is considered as the best equation to estimate the non-marine background sulphate concentration ($SO_{4\,o}^*$) in Sweden.

The role of organic matter has not been possible to evaluate and therefore not included in the calculation.

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