

The Sulphur Budget of Sweden During this Century

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Based on data from the Swedish water quality covering 78% of the Swedish area, the discharge of sulphur has been computed for the period 1969 to 1977. The trend for this period gives a figure for 1977 of 660,000 tons per year. This figure exceeds that of the atmospheric fallout by about 200,000 tons per year. The different items contributing to this difference are discussed.

The increase of the discharge of sulphur during the investigated period varies from 2 to 5% per year in the different regions of Sweden. Due to the storage characteristics of the soil-water reservoirs the increase of the input of sulphur must be larger than these figures.

The discharge of sulphur from different parts of Sweden during this century has been reconstructed. The total figure in excess of a geochemical reference amounts to about 60 million tons of sulphur. In parts of the South and West of Sweden the annual deposition of sulphur amounts to about 25 kgs per ha. Rough figures of the so-called biogenic sulphur can be given for Sweden along with a geochemical reference value.

The application of these figures to the increasing acidification of Sweden is obvious.

Introduction

The regional acidification of Europe (Odén 1968, 1975) has now been widely accepted as being mainly caused by the anthropogenic emissions of sulphur into the atmosphere (OECD 1977). Consequently, the extent of the anthropogenic acidification is closely related to the fallout of sulphur, and much work has been

devoted to determinations (wet fallout) and computations (dry fallout) of the total fallout of sulphur from meteorological and atmospheric chemical data. The problem has been thoroughly discussed at the First International Symposium on Acid Precipitation and the Forest Ecosystem (Dochinger and Seliga, Eds. 1975), one year later in Telemark, Norway (Ambio 1976), and most recently at the Symposium on the Effects of Acid Precipitation on Terrestrial Ecosystems in Toronto (Hutchinson, Ed. 1979). The atmospheric chemical aspects of the problem with special reference to sulphur were discussed at the international Symposium on Sulphur in the Atmosphere in Dobrovnic (Husar, Ed. 1977).

The total fallout of sulphur over Sweden during the last decade has been estimated by different authors, Table 1. The figures do not vary materially. Nevertheless, there is some uncertainty of the total deposition of excess sulphur mainly due to the difficulties in the computations of the amount of the dry deposition. Furthermore, there is an inherent error in the computations of the fallout of sulphur, which is due to looping (Odén 1975). This process means that sulphur is recycled between the vegetation-soil-water reservoirs and the atmosphere and vice-versa. Depending on the intensity of looping, the measured fallout of sulphur will exceed the net fallout of sulphur. The figures given in the table are consequently maximum figures.

As yet, the consequences of looping has not been considered with respect to the fallout from the atmosphere. Consequently, the computations of global budgets are fictitious to a certain degree. Eriksson (1963), however, has been aware of the effect of looping in his studies on the circulation of sulphur in Nature.

A determination of the total net fallout of sulphur can also be made from the discharge of sulphur by the rivers. This figure is a maximum figure with respect to the atmospheric fallout and corrections have to be made for the industrial pollution of sulphur into the rivers, sulphur due to weathering and the agricultural use of sulphur in fertilizers. The amount of sulphur originating from the ocean

Table 1 - Deposition of total excess sulphur² over Sweden according to different authors.

Year	Ref.	Deposition of S	
		kg · ha ⁻¹ · yr ⁻¹	ton · yr ⁻¹
1965	Bolin et al., 1971	10.4	464,000 ¹
1974	OECD, 1977	10.0	447,000
1976	Granat, 1978	11.2	500,000

1. The figure has been computed by the author assuming an addition of sulphur due to dry deposition by 75% and 50% in Southern and Northern Sweden respectively, in relation to the wet deposition.
2. Equals total sulphur minus the contribution of marine sulphur.

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water has also to be accounted for. By means of existing data the following computations have been made:

- 1) The discharge of sulphur from Sweden during 1969-1977.
- 2) The transient parts of the discharge of sulphur during 1965 to 1977.
- 3) Reconstruction of the anthropogenic part of the Swedish sulphur budget during this century.
- 4) Estimates of geochemical reference values and the probable values of biogenic sulphur before the industrial period.

Total Discharge of Sulphur by the River Systems

In 1961 a preliminary water chemical network was initiated in Sweden and Finland (17 river basins) with a limited analytical program (Odén 1964). In 1965 this network was supervised by Ahl, who successively extended the network to 73 river basins (Odén and Ahl 1970; Ahl and Odén 1972). Water samples are taken every month and more than 20 different determinations are made on each sample. Part of the data from this network is used in this paper.

The drainage area covered by the network is given in Fig. 1 along with the appropriate network stations. With the specific purpose to obtain figures of the total discharge of sulphur from Sweden, only the stations closest to the outflow of the rivers to the Baltic Sea or the North Sea have been utilized. To simplify the presentation, Sweden has been divided into five regions, which are more or less homogeneous with respect to hydrological and atmospheric chemical conditions. Altogether data from 36 sampling stations have been used and they cover 78% of the area of Sweden. The variations in this respect for the different regions are given in Table 2. To illustrate the long-term effects some of the stations starting in 1961 have also been utilized.

Some river systems extend outside Sweden, altogether 24,700 km² or 5.2% of the total drainage area. Consequently, the discharge of the different elements have been corrected for this external area by means of the discharge coefficient for the appropriate river system. On the other hand, 22% of the land area is not incorporated in the network. The discharge contribution from this area is computed from the mean discharge coefficient for each region.

The data from the Swedish water quality network show, that the discharge of an element may vary from year to year. Consequently, long-term data (»10 years) are necessary in order to obtain a trend in the discharge of any elements. There is, however, a simple method to cancel these yearly variations. This is based on the fact, that the variation in the discharge of an element is basically related to the discharge of water. In a mechanistical sense this means that the yearly discharge of

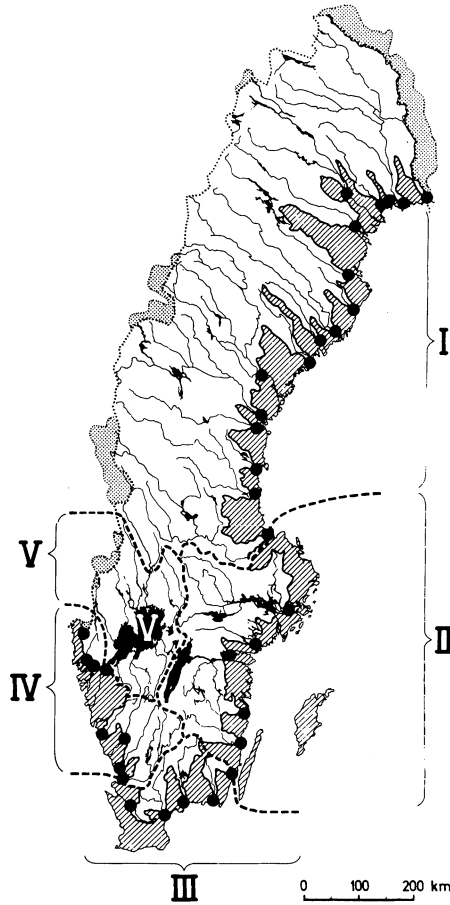


Fig. 1. Map of the Swedish water quality network. The network stations used in this study is denoted by dots. Drainage areas outside Sweden is denoted by \otimes , areas not incorporated in the network by $///$. Sweden has been divided into five regions, I-V.

an element ($c_i \cdot q_i$) “drains” only a fraction of a large reservoir of that element within the drainage area. The model for eliminating the yearly variation of the fluxes of elements is, consequently, to eliminate the variation in the discharge of water, or

$$c_i q_i \left(\frac{\bar{q}}{q_i} \right) \rightarrow c_i \bar{q}$$

actual
expected
discharge
discharge

where \bar{q} means the average discharge coefficient of water during the measuring

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Table 2 – Data on the regions used in the computations of the discharge of sulphur from Sweden. The five regions are illustrated in Fig. 1.

Region	No. of rivers	Area, km ²			Discharge correction factor
		total ¹	studied ³	%	
I	18	290,100	240,200	83	1.21
II	5	69,100 ²	46,900	68	1.47
III	5	23,800	9,700	41	2.46
IV	7	28,700	15,800	55	1.82
V	1	36,300	36,300	100	1.00
Total	36	448,000	348,900	78	not used

1. Areas within Sweden.

2. Including Öland and Gotland.

3. Corrected for areas outside Sweden. For region I 22,800 km², for region IV 1,900 km².

period. Since the drainage basins are not perfect, well-mixed reservoirs, actual and computed discharges of an element are not identical over a period of years, or in other words

$$\sum_{1969}^{1977} c_i q_i \neq \bar{q} \sum_{1969}^{1977} c_i \quad \text{OR} \quad \sum_{1969}^{1977} c_i q_i = \delta \bar{q} \sum_{1969}^{1977} c_i$$

By testing the above “identity” it was found, that the computed discharge of sulphur differed from the actual figure by $\pm 5\%$ ($\delta = \pm 1.05$).

In order not to distort the actual data quantitatively, the discharge of sulphur from each region has been corrected by the δ -values appropriate for each river.

The difference between the actual discharge of sulphur and the computed values based on ‘constant q ’ are shown in Fig. 2. Actual data vary considerably from year to year and possible time trends are statistically insignificant. On the other hand, the corrected figures show little variation between years, and the trends are statistically significant (except Råne älv). The residence time of water in the drainage areas vary from approximately 50 years (lake Vättern) to 3 years (Råne älv). The residence time of sulphur is larger than the residence time for water (Odén 1979).

Considering Sweden as a whole the actual discharge of sulphur varies considerably from year to year, especially in the latter part of the period. In 1976 the figure was only 460,000 tons but increased to 830,000 tons the year after. The q -corrected values are smoothed and increase by about $2\% \cdot \text{yr}^{-1}$. The smoothed value for 1977 amounts to 660,000 tons which is much less than the actual figure.

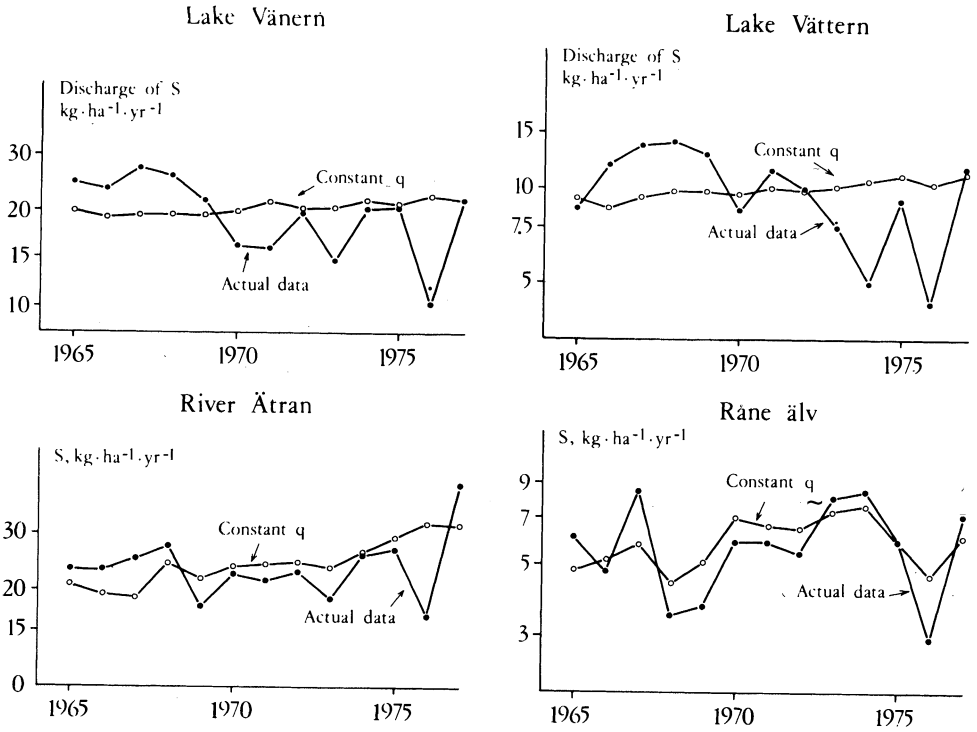


Fig. 2. The yearly discharge of sulphur from four river systems. Dots denote actual data, circles those corrected for constant Q (see text).

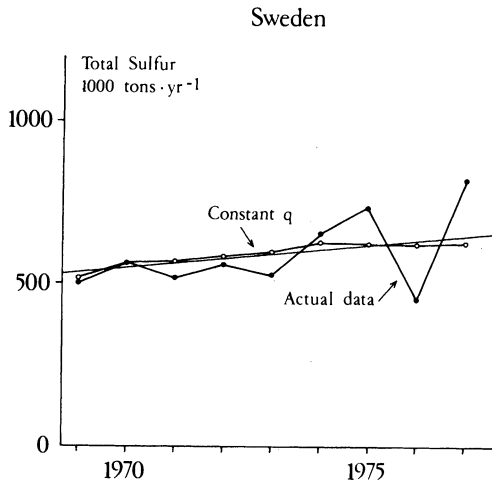


Fig. 3. The same as for fig. 2 but related to the whole of Sweden.

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In 1974 the situation is reversed, which has been commented on in a previous paper (Odén and Ahl 1979).

The analytical procedure behind the determinations of the total discharge of sulphur by the rivers does not include sulphur in organic matter (dissolved humus). The sulphur content in different humus fractions varies between 0.4 and 0.7%. The mean value for the soluble fractions is about 0.5%. The total discharge of organic matter has been reported to be 1,440,000 tons per year (Ahl and Odén 1972). This figure amounts to about 2,000,000 tons when the total drainage area of Sweden is taken into account. The discharge of S in the organic matter can thus be computed to about 10,000 tons per year. Most of it is released from region I. There are no transient conditions of this form of sulphur, and if any, it is likely to be negative, since the amount of dissolved humus is shown to be reduced with time (Ahl 1979). This may be due to the flocculating effects originating from increased acidity of the river waters and the increased concentration of divalent cations.

The probable discharge figures in 1977 from the different regions are given in Table 3. For all regions except No. I, the discharge coefficient is larger than 21 kgs S · ha⁻¹ · yr⁻¹. Some drainage basins show much higher figures. Thus the discharge from Lake Mälaren is as high as 41 kgs S · ha⁻¹ · yr⁻¹ (the highly industrialized Mälär region). The value for Rönne å in Skåne is almost 54 kgs S · ha⁻¹ · yr⁻¹. On the other hand the lowest figure appears from Råne älv, 6 kgs S · ha⁻¹ · yr⁻¹.

The rate of change of the discharge coefficient (in kg · ha⁻¹ · yr⁻¹) is also given in Table 3. The relative and the absolute figures are lowest in N Sweden. The

Table 3 – The total discharge (1977) and the rate of change of the discharge of sulphur during the period 1965-1977.

Region	Total discharge of S., tons·yr ⁻² 1	Disch. coeff. kg·ha ⁻¹ ·yr ⁻¹	Sulphur trend		
			No. of rivers	%·yr ⁻¹	kg·ha ⁻¹ ·yr ⁻¹
I	242,000	8.3	7	+1.7	+0.15
II	195,000	28.2	4	+3.1 ²	+0.87
III	64,000	26.9	4	+3.7	+1.02
IV	72,000	25.1	2	+3.9	+0.97
V	75,000	20.7	1	+1.2	+0.26
Sum or mean	648,000 ¹	14.5 ³	18	2.1 ³	0.56 ³

1. 10,000 tons of S originating from S in organic matter is not included.
2. The figure for Lake Vättern is only 1.6% per year.
3. Weighted averages.

percentage increase is also very low from unpolluted drainage basins with large water volumes like lake Vättern and lake Vänern. The total increase of the discharge of sulphur amounts to 25,100 tons per year. When the effects of fertilizer-S also are taken into account (cf. below), this figure increases to 26,600 tons per year. Since the effects of weathering and marine salt over so short a period as 10 years are negligible, this accelerating input of sulphur over Sweden must be due to increased anthropogenic emissions in Sweden and the rest of Europe. However, the Swedish measure to reduce the domestic emissions have not substantially reduced the fallout within Sweden. This was also to be expected, since the sulphur sources outside Sweden account for about 80% of the fallout within Sweden (OECD 1977).

Sulphur through Weathering

In ordinary bedrock and soil material the sulphur content is about 0.05%. Some of this sulphur is released by weathering of the mineral particles. This release was studied in 48 soil samples from 12 representative sites from the North to the South of Sweden (*A*₂, *B* and *C* horizons of iron podsoles). The weathering was made step-wise in a geometric time progression during one year using 0.1 M HCl. During the time of such an artificial weathering, 3% of the original material dissolved at a maximum. Totally 15 elements were determined. (Odén and Bergholm 1979).

Some results are given in Fig. 4. The release of sulphur from the Middle and North of Sweden amounts to about 2 g per kg weathered products and there is almost no change in the release of sulphur with increasing weathering intensity. Along the West coast of Sweden the release is higher and increases with increasing weathering intensity. The 6 samples from Skåne and Småland show a pattern

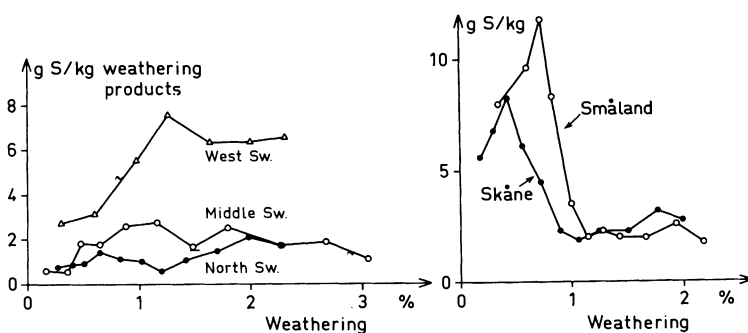


Fig. 4. The release of sulphur by successive acid weathering of soils from different parts of Sweden.

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which suggests that some S-rich, easily weatherable minerals occur in these parts of Sweden. Above 1% weathering, the release of sulphur is reduced to the same range as for the other areas in Sweden. The S-rich material is consequently a limited S-source in these areas.

Table 4 gives the release of sulphur due to weathering. The computations are based on the total discharge of silicon by the rivers and the S/Si ration of the weathering products relevant for the different regions. The computed figures are very low and less than 0.7% of the total discharge of sulphur. Even if some silicon is trapped in the soil (as amorphous SiO₂) or in the lake basins (as diatom SiO₂) the contribution to the sulphur budget due to weathering is almost negligible. Neither will increased weathering increase the discharge of sulphur to any measurable extent.

Table 4 - Computed figures for the release of sulphur due to weathering.

Region	Discharge of Si	Sulphur due to weathering	
	tons·yr ⁻¹	tons·yr ⁻¹	kg·ha ⁻¹ ·yr ⁻¹
I	190,600	900	0.03
II	36,900 ¹	900	0.13
III	13,400	800	0.34
IV	21,400	1,000	0.35
V	30,000 ¹	800	0.22
		4,400	

1. Corrections have been made for Si trapped in the Great Lakes.

Marine Sulphur

Some of the sulphur in the river systems is due to cyclic salts originating from the oceans. It is possible to compute the amount of cyclic sulphur from the content of sulphur of ocean water in proportion to that of sodium, chloride or any other conservative element. We have used sodium since only small amounts of sodium is released by chemical weathering (cf. Table 5). Sodium makes up only 2% of the released amount of silicon as found by the weathering studies. Chloride is somewhat hazardous because the frequent use of CaCl₂ as a road salt during the winter period.

The data for the different regions are given in Table 5. Marine sulphur amounts totally to 37,800 tons, which is about 6% of the total discharge of sulphur by the

Table 5 – Computed figures of the contribution of marine sulphur for the period 1969-1977 based on the discharge of Na.

Region	Discharge of Na	Marine sulphur ¹	
	tons·yr ⁻¹	tons·yr ⁻¹	kg·ha ⁻¹ ·yr ⁻¹
I	167,000	13,700	0.47
II	84,000	6,900	1.00
III	63,000	5,300	2.23
IV	64,000	5,300	1.85
V	80,000	6,600	1.82
Sum		37,800	

¹ Calculated from $S_{\text{marine}} = 0.084 (\text{Na} - 0.02 \text{ Si})$ where $0.084 = \frac{S_{\text{marine}}}{\text{Na}_{\text{marine}}}$ and 0.02 Si equals the contribution of Na in the river systems due to weathering. The coefficient 0.02 is based on experimental studies on different Swedish tills.

ivers. The discharge per unit area varies with the different regions and reflects the distance from the North Sea. The figures for region II and V seem to be too high. This is probably due to the land rise, which leads to successive exposure and drainage of fossile salt water from the glacial period and to the cultivation of the land, which releases sulphur from acid sulfate soils.

Since the atmospheric fallout of sodium and chloride has been constant during the last 20 years there is no reason to believe, that the amount of marine sulphur is in a transient stage. Consequently, the contribution of marine sulphur is assumed to have been constant during this century.

Sulphur in Fertilizers

Sulphur is added to cultivated areas by means of different fertilizers such as ammonium sulfate (24% S), potassium sulfate (18% S) and superphosphate (14% S). From available statistics it is possible to compute the input of fertilizer-S to the cultivated areas in Sweden during the last 70 years. As can be seen from Fig. 5, the figure is about 7 kgs S/ha and year up to 1940 and increases to more than 20 kgs S/ha and year during the 1950's. This is mainly due to the increased use of superphosphate and ammonium sulfate. Up to the present time there is a gradual decrease of the input of sulphur amounting to approximately 0.5 kg per ha and

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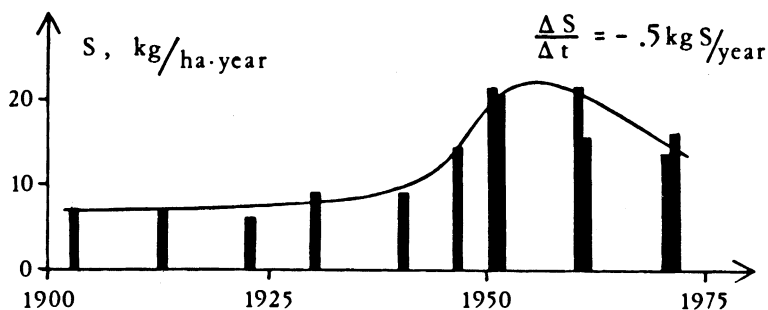


Fig. 5. The consumption of fertilizer sulphur in Sweden during this century. The figures are related to the agricultural area. Corresponding figures for the different regions in 1970-72 are given in Table 6.

year. Since the sulphate ion is not firmly bound to the soil colloids, this negative trend will also influence the water chemistry especially from more intensively cultivated drainage areas.

In 1970-72 the total use of fertilizer-S amounted to 44,300 tons/year. The net effect of this input of fertilizer sulphur on the discharge of sulphur by the rivers will, however, be less than the above figure since part of the added sulphur will escape to the atmosphere as hydrogen sulphate or as dimethyl sulphide. This is part of the mechanism denoted by looping. The distribution between the different regions is given in Table 6 along with computed figures for the rate of change. The figures are fairly low when plotted on bases of the drainage area of the different regions, and small variations from one year to another do not materially interfere with the budget figures for Sweden.

Table 6 - The input and the rate of change of the input of fertilizer sulphur of the five drainage regions of Sweden.

	Fertilizer-S	Rate of change
	kg·ha ⁻¹ ·yr ⁻¹	kg·ha ⁻¹ ·yr ⁻¹
I	0.3	-0.1
II	2.3	-0.07
III	4.0	-0.13
IV	1.5	-0.5
V	1.1	-0.1

The Net Discharge of Sulphur During This Century

By means of the preceding data it is possible to compute the net discharge of sulphur by the rivers from the different regions of Sweden, Table 7. The total figure amounts to 578,000 tons of S in 1977. As seen in Table 7, some corrections have to be made for the transient part of fertilizer-S from 1971/72 to 1977, and for sulphur in organic matter transported by the rivers. If we assume this amount of sulphur to be caused by atmospheric fallout (or in part being generated within the soil-water system) comparison between input and output at a given time can be made only if we consider the soil-water reservoir being in a transient stage. As shown by the data in Table 3 the change of the discharge of sulphur is increasing with time. This means the input must increase too and, furthermore, always be larger than the output. Experimental data show that the rate of change $\partial f_0 / f_0 \partial t$, is constant ($k_m(\text{measured})$) but not equal for the different regions and varying from +1.7% to +3.9% per year. The outflow (f_0) is made up by a constant part due to the geochemical background (f_0^b) and another part due to the transient discharge of anthropogenic sulphur (f_0^{tr}). In a paper of transient reservoirs (Odén 1979) the following has been shown.

The rate of change of the transient flux of sulphur k_{tr} is related to the measured rate of change of the discharge of sulphur by

$$k_{tr} = k_m \frac{f_0}{f_0^{tr}}$$

Table 7 – The discharge of sulphur in 1977 from the different regions of Sweden.

	Discharge coefficient kg S·ha ⁻¹ ·yr ⁻¹	Sum of contributions ¹ kg S·ha ⁻¹ ·yr ⁻¹	Anthropogenic-S plus geochemical background	
			kg S·ha ⁻¹ ·yr ⁻¹	tons·yr ⁻¹
I	8.3	+0.8	7.5	217,000
II	28.2	+3.4	24.8	171,000
III	26.9	+6.6	20.3	48,000
IV	25.1	+3.7	21.4	61,000
V	20.7	+3.2	17.5	64,000
Sum for the whole of Sweden			561,000	
Fertilizer-S 1972 → 1977 ²			+ 7,000	
Organic-S in humus			+ 10,000	
Discharge (= Fallout)			578,000	

1. Weathering-S, marine-S and fertilizer-S
2. The change for the period

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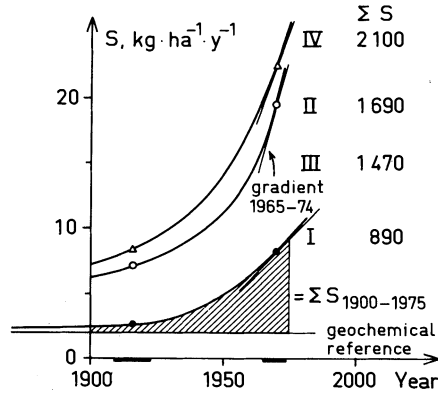


Fig. 6. The probable development of the discharge of anthropogenic sulphur for region I, II and IV during this century. The gradients for the period 1965-1977 are denoted as well as the probable geochemical reference for region I. ΣS denotes the accumulated discharge of sulphur per ha during this century.

Furthermore, the transient part of the influx, f_i^{tr} , in relation to that of the discharge of sulphur is given by

$$f_i^{tr} \equiv f_0^{tr} (1 + k_{tr} \tau_s)$$

τ_s denotes the residence time of sulphur in the soil-water reservoir. τ_s is always larger than τ for water, which is transporting the sulphur (Odén 1979). The above expression holds also for derivatives and integrals of the fluxes. Finally, the difference in time ($t_i - t_0$) when the fluxes are equal is constant and given by

$$t_i = t_0 = \frac{\ln(1 + k_{tr} \tau_s)}{k_{tr}}$$

In the present problem the rate of change of the discharge of the transient part of sulphur is approximately 30% larger than the measured values. This means that the rate of change of the influx of sulphur from the atmosphere (or generated within the soil-water system) is about +5% per year in Sweden south of Latitude 60°N. This figure can also be computed from the atmospheric chemical data from Scandinavia during the period 1952 to 1970 (Granat 1972). This is a strong support to the view that the increase in the discharge of sulphur by the rivers originates from atmospheric fallout.

In order to generate the observed discharge conditions during the last decade the influx f_i^{tr} must have been some 20% larger than the discharge f_0^{tr} at any

time during this decade. This means that a discharge of 578,000 tons of sulphur in 1977 must be caused by an influx of 668,000 tons of sulphur. The difference between the figures equals the increased storage of sulphur in the soil-water reservoir in 1977. The figure of 668,000 tons of sulphur is much higher than the fallout computed from atmospheric chemical data. Actually, there is a difference between expected atmospheric fallout of sulphur and the measured/computed fallout of sulphur of almost 200,000 tons (668,000 minus about 500,000, Table 1). This difference, however, is still larger when the effect of looping is considered. We believe that this difference is real and may arise from any one of the following causes to a varying degree.

- 1) The atmospheric chemical network does not account for all forms of sulphur such as elemental sulphur.
- 2) Sulphur in soil organic matter is in a transient stage due to increased mineralisation.
- 4) Sulphur in marine sediments and in acid sulfate soils are continuously being released.

Because of lack of data from a soil chemical network it is not possible to evaluate the magnitude of the different sources of the unaccounted amount of sulphur found in the river waters. Some principle results, however, are given in the paper by Odén (1979).

The net atmospheric deposition in excess of marine salts is made up of two parts: one originating from anthropogenic emissions, the other due to the non-anthropogenic background (sulphur from volcanoes, global dust, biogenic sulphur etc.). It is to a certain extent possible to evaluate the two parts by means of old Swedish data, and furthermore, to describe the progression of the fallout of sulphur due to anthropogenic emissions during this century.

The situation is as follows: The rate of change of the discharge of sulphur from the different regions in Sweden can be reconstructed by use of the following facts: the mean discharge of sulphur during the period 1965-1977, the rate of change of the discharge of sulphur during the same period, and the discharge of sulphur for various years during the period 1909-1923. The latter data have been given by J. V. Eriksson (1929) and have been recomputed with respect to the five regions.

The results are given in Fig. 5. The curved lines show the probable change of the discharge of sulphur from 1900 to the present time satisfying the three conditions mentioned above. From the curves it is clear, that the increase of the discharge of sulphur started before 1900 and that the discharge has increased 3 to 4 times in 75 years. A similar increase (2 to 3 times) has also been found in the Greenland ice cap during the corresponding period. The total, excess discharge of sulphur for the four regions amounts to the figures given in Fig. 5. According to what has been stated before the accumulated fallout of excess sulphur must be about 20% larger.

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The non-anthropogenic background is only indicated in Fig. 5. The figures vary from about 2 kgs sulphur per ha and year (region I) to 5-8 kgs for the other regions. This means that the fallout of anthropogenic sulphur amounts to 6 to 18 kgs of S per ha/year in 1977. Going into the details within the different regions these figures vary still more. In the most southern part of Sweden the anthropogenic contribution is likely to be around 25 kgs S per ha/year.

Acknowledgement

This work has been supported by the Natural Science Foundation of Sweden, contract No. G 3887-002, and the Research Council of the Swedish Environmental Protection Board, contract No. 7-142. The author is much indebted to Dr. T. Ahl for his permission to use the data from the Swedish water quality network.

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Received: 14 February, 1979

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