A study of the sulfur budget for the atmosphere over Northern Europe[1]

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ABSTRACT

The atmospheric sulfur budget for Northern Europe is discussed and compared with global averages. It is found that in this area the anthropogenic sulfur sources outweight natural sources. A comparison between deposition and emission of sulfur within different regions in Northern Europe shows that the dispersion of sulfur has a continental character; i.e. since, on average, sulfur is transported more than 1 000 km before it is deposited, one country can be appreciably influenced by another, in this respect. A tentative model for the variation of deposition with distance from a source is suggested. Application of this model gives an estimated turnover time for anthropogenic sulfur of 2–4 days. As a quantitative example, the distribution between various sources of the sulfur deposition in Sweden has been estimated. The calculations indicate that about half of the sulfur originates from foreign anthropogenic emissions, whilst the other half is caused by Swedish emissions and a natural background. Just as Sweden is influenced by the surrounding industrial countries, it is furthermore clear that a large part of the sulfur emitted in Sweden is deposited outside the country.

1. Introduction

The present study was undertaken as a contribution to a case study ("Air pollution across national boundaries; The impact on the environment of sulfur in air and precipitation", Stockholm 1971), which has been prepared for the UN conference on the human environment 1972. It covers not only meteorological and chemical aspects of sulfur as an air pollutant, but also biological, technical and economical problems. In this paper we confine ourselves to a discussion of some of the meteorological aspects.

When discussing sulfur as an air pollution problem it is important to recognize that we deal with basically two different problems, even if they are interconnected and sometimes difficult to distinguish. The first is the problem of the possible damage (corrosion, health effects, damage to vegetation) caused by the SO_2-gas—and often other pollutants at the same time—in the air. Since such effects probably do not occur for concentrations below a few parts per hundred million (by volume), and since higher concentrations, at least in Scandinavia, are found only in the neighbourhood of local SO_2 sources, such damages is essentially a local problem, and thus distant sources are unimportant. This problem will not be discussed further there.

The other aspect of sulfur as a pollutant and the one which we are concerned with here, is the possible damage caused by sulfur deposition. In this case, it is the accumulated amount of acid deposited on the ground that is important. SO_2 and H_2S in the atmosphere are oxidized and eventually transformed into sulfuric acid. If this acid is not neutralized by other compounds in the air, it increases the acidity of rainwater. An addition of acidified precipitation to soils and waters may cause damage to biological life. Although oxidized sulfur is not the only compound determining the acidity of the deposition, the observed increase in sulfur deposition over some areas in Northern Europe (Munn & Rodhe, 1971) probably is the main factor causing the observed increase in the deposition of acid. Some aspects of the balance between acids and bases in the atmosphere, and also the observed deposition patterns for sulfur and acid, is discussed separately (Granat, 1972*a* and *b*).

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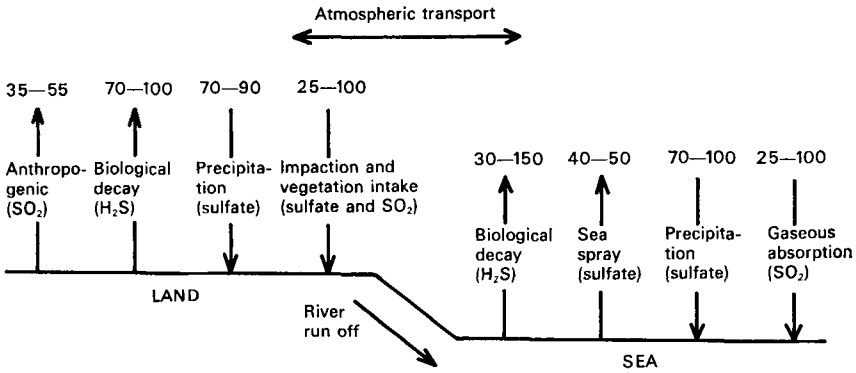

Fig. 1. The global atmospheric sulfur budget around the year 1965. Unit: Million tons sulfur annually.

Since an increased acidity of precipitation has been measured over large areas in Northern Europe, not only in the neighbourhood of strong SO_2 sources, it seems clear that the deposition of sulfuric acid is not only a local problem, but that the influence of distant sources needs to be considered. The purpose of this paper is to study the dispersion and deposition of anthropogenic sulfur. The question of the extent of influence of a sulfur source, is closely connected with the question of the turnover time of sulfur in the atmosphere. We have therefore tried to estimate such a time scale for anthropogenic sulfur in the atmosphere over Northern Europe by making certain budget calculations; i.e. we have compared the deposited amounts with the emissions within different regions. As an example of the influence from distant sources we also try to estimate the relative importance of the contributions from Swedish and non-Swedish anthropogenic sulfur sources for the sulfur deposition in Sweden.

2. Estimates of the atmospheric sulfur cycle

On the basis of available data, Eriksson (1960) discussed the circulation of sulfur in nature. Estimates of the global sulfur budget can also be found in a paper by Eriksson (1963) and have also been given by Junge (1963). A more recent attempt to review some of the underlying data has been made by Robinson & Robbins (1970). Fig. 1 shows the principal sources and sinks for atmospheric sulfur and estimates of their strengths. The figures are, by and large, taken from the above mentioned papers and the intervals reflect the uncertainties. It is evident that the anthropogenic emissions represent a significant addition to the natural cycle but that, in a global balance sheet, they are still smaller than the natural emissions. Since sulfur has a turnover time in the atmosphere which is short compared to the time for mixing of the troposhpere, it is also relevant to study the sulfur budget within limited regions of the atmosphere. Due to the non-uniformity in the geographical distribution of the sulfur sources, such considerations may result in a picture quite different to that of Fig. 1 (cf. section 3).

Meetham (1950) studied the budget of SO_2 over England and obtained a turnover time for this gas in the atmosphere of about 10 hours. Junge (1960) re-examined this estimate and found that the turnover time might be a few (4) days. In the same paper, by comparing the emissions with the deposition over the continent and estimating an average horizontal transport velocity, Junge estimated a turnover time for anthropogenic SO_2 in the U.S.A. of 7 days. A similar study but on a smaller scale was carried out by Rodhe (1970) in the surroundings of a Swedish city. It was found that only a very small part of the anthropogenic sulfur from the city was deposited within the nearest 10 km and that the turnover time for this sulfur must have been *at least* 5 hours.

3. Sources and sinks for atmospheric sulfur in Northern Europe

In the present study some regions of different sizes in Northern Europe are considered. In

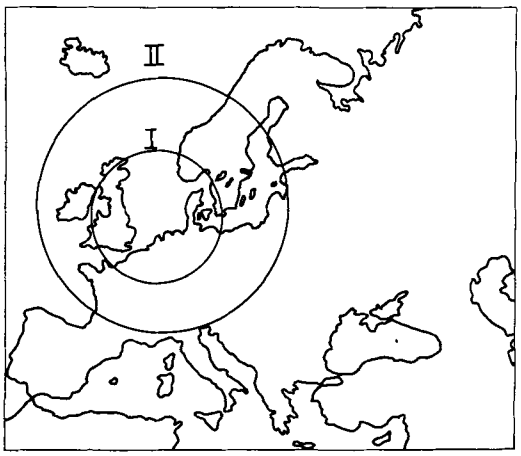

Fig. 2. Definition of the two regions used for the sulfur budget calculations.

Fig. 2, two of the regions to be used in the following discussion are defined.

Region II, covers approximately 1 % of the earth's surface and within this region the anthropogenic sulfur emissions are about 10^7 tons S/year (for further details see below). If the natural sources were roughly uniformly distributed over the earth's surface—there is no strong evidence that they are not—the natural emissions within the same region would amount to only $1–2 \times 10^6$ in the same units. This does not include sulfur in sea salt spray. This sulfur is neutralized and does not contribute to the acid load. The amount of sea salt sulfur is easily estimated from measurements of Na or Cl and can therefore be subtracted from the total amount to yield the "excess sulfur". Only excess sulfur will be discussed in the following sections. For region II, and even more so for region I, the anthropogenic sulfur emission thus most likely dominate over the natural emission. This conclusion is supported by a study of the chemical composition of river waters performed by Berner (1971).

A rough picture of the distribution of anthropogenic sulfur emission in some countries in Northern Europe is shown in Table 1. For some of the subsequent calculations a further subdivision between the emissions in different regions has been made.

The most important processes that remove the sulfur from the atmosphere are (cf. Fig. 1):

(*a*) *Washing out by precipitation* (both SO_2 and particles containing sulfate) *and sedimentation of particles containing sulfate.* The amounts deposited by these processes are measured at several places and are thus rather well known (Eriksson, 1970; Granat, 1972*b*).

(*b*) *Impaction of particles containing sulfate on the ground or on vegetation.* From the balance sheet between input from the atmosphere and output through river run off, Eriksson (1955) estimated that in Sweden the amount of deposition of chlorides by this process was about twice as large as that through precipitation. Investigation by White and Turner (1970) showed similar results for chloride and other sea-salt aerosols in a woodland canopy. On the other hand, for calcium and phosphorus which have different sources, these authors estimated that the dry deposition was less than that through precipitation.

(*c*) *Absorption of SO_2 by the vegetation, soils and waters.* Although there is some information from laboratory measurements, it is difficult at the present time to estimate the amounts of SO_2 that are absorbed. It seems, however, that within the regions considered this process is less significant than the precipitation scavenging.

Based on the above considerations it is estimated that the total deposition of anthropogenic sulfur in Northern Europe is between 1 and 2 times the precipitation deposition.

Fig. 3 shows the geographical distribution of the annual precipitation deposition of sulfur. The values are nominally those for the year

Table 1. *An estimate of the anthropogenic emissions of sulfur compounds from a number of countries in North-Western Europe 1968 (OECD figures)*

The values refer to emissions from the burning of fossil fuels. The total emissions (including industrial processes) are somewhat (appr. 10 %) higher

	10^6 tons S/year
Belgium, France, Germany, Luxembourg, Netherlands	3.7
Austria, Switzerland	0.2
Scandinavia	0.6
United Kingdom, Ireland	3.1
Neighbouring countries in Eastern Europe	1–3 (here estimated)

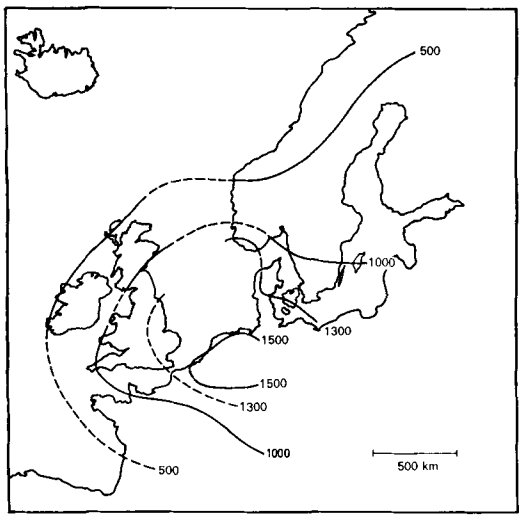

Fig. 3. Deposition of sulfur through precipitation during one year (average for a few years around 1965) expressed in mg sulfur/m² (Granat, 1972*b*). The sulfur originating from sea spray has been deducted.

1965, but are, in fact, average values for a few years around this time. The deposition values apply to areas outside the immediate vicinity of the built-up areas. The deposition within small areas in the vicinity of the sources can be a few times larger than the values given (Andersson, 1969; Rodhe, 1970). The contribution of this increment to the total amount of sulfur deposition within larger areas is, however, probably insignificant.

Since we want to compare the sulfur deposition with the anthropogenic emissions, it is important to know what part of the deposition that can be ascribed to these emissions and what is a natural background deposition level. This background level, which in this case corresponds to the deposition that would occur if there were no anthropogenic emissions in Europe, can be roughly estimated in the following way:

The total amount of precipitation deposition for the earth as a whole is, according to Fig. 1, about 160×10^6 tons S/year. From this figure we subtract the sea spray (~ 45) and the European anthropogenic sulfur (~ 10). If the rest were uniformly distributed over the earth's surface, the annual deposition would be 210 mg S/m². In view of the fact that one may expect a tendency for the natural sulfur sources—and thereby also to some extent the sinks—to be located on land and in coastal areas, this figure is probably somewhat low when considering the

Table 3. *The sulfur budget for some countries in Northern Europe for the year 1965. (Unit: 10^6 tons S/year.) Cf. Table 2*

Country	Anthropogenic sulfur emission	Deposition by precipitation minus an estimated background level	Total deposition as % of anthropogenic emission
S. Sweden (south of lat. 61° N)	0.23	0.13	60–110
N. Sweden	0.07	0.09	130–250
Norway	0.08	0.10	125–250
Denmark	0.12	0.05	40–85
Holland	0.5	0.05	10–20
W. Germany	1.6	0.30	20–35
France	1.1	0.25	25–45
U. Kingdom	3.2	0.25	10–15

Table 2. *Sulfur balance sheet for two regions in Northern Europe for the year 1965 (Unit: 10^6 tons S/year)*

The intervals in the last two columns stem from the fact that the total deposition may be up to twice as large as that through precipitation only

Area (cf. Fig. 2)	Anthropogenic sulfur emissions	Deposition of sulfur by precipitation (cf. Fig. 3)	Deposition by precipitation minus an estimated background level	Total deposition, including other processes than precipitation	Total deposition as % of anthropogenic emission
I	5.9	1.5	1.2	1.2–2.3	20–40
II	8.4	4.5	3.2	3.2–6.4	40–75

deposition in Europe. The lowest values measured in remote areas in Northern Scandinavia (cf. Fig. 3) are approximately 300 mg S/m² annually. We thus estimate the natural background level, defined as above, in Northern Europe to be about 250 mg S/m² annually.

In Table 2 a comparison has been made between the deposition of sulfur as computed from the pattern in Fig. 3 and the anthropogenic emission within the two regions shown in Fig. 2. Even if these calculations are uncertain, they do indicate that within the inner region (I) the quantity of sulfur deposited is less than one-half (20–40 %) of the quantities emitted within the same region. Even for region II a considerable part of the emission is quite clearly exported to regions outside the area.

Another way of illustrating that, in many cases, one country is not independent of others, is to look at the sulfur budget for individual countries, i.e. compare the amounts emitted with what is deposited within each country. The result of such a comparison for some countries in Northern Europe is shown in Table 3.

4. A model of the variation of deposition with distance from the source

The average concentration of sulfur in the air, and thereby also the deposition of sulfur per unit area on the ground, decreases with increasing distance from a source. This is due to the dispersion of the sulfur over a larger area and to the depletion itself. Since we are not dealing with the pollution situation on a particular day, but with the mean value over a period of the order of years, it is not the dispersion from a single plume that is relevant. Rather, we are concerned with the effect of the synoptic scale motions and we can study the spatial dispersion by looking at the end point of a large number of air trajectories released at different times from the source. (Cf. Durst et al., 1959.)

Let us, for a moment, assume that the distribution of such trajectory end points is symmetric around the source, i.e. all transport directions have the same probability. In this case it is possible to describe the deposition by a function depending only on the distance r from the source. This function should include a factor $1/r$ reflecting the decrease in deposition corresponding to the increase in area when r is increased. The depletion caused by the deposition itself can be taken into account by multiplying with an exponential factor. We thus suggest the following approximate form for the deposition function (valid outside the immediate vicinity of the source)

$$d(r) = \frac{A}{r} e^{-r/r_0} \tag{1}$$

where $d(r)$ is the deposition per unit area and unit time, and A and r_0 are constants.

The constant r_0, which determines the decay rate, is related to a turnover time τ by the equation $r_0 = V\tau$, V being the average transport velocity from the source.

The constant A can be determined by the condition that the emitted amount M (per unit time) should equal the total deposition (per unit time)

$$M = \int_0^\infty 2\pi r\, d(r)\, dr = 2\pi A r_0 \tag{2}$$

Thus

$$A = \frac{M}{2\pi r_0}$$

As noted above, eq. (1) cannot be valid very close to the source ($r \leqslant r_1$). However, when studying the total deposition within a distance r from a source, $D(r)$, over a large area, $r \gg r_1$, the error introduced by using an incorrect $d(r)$ for $r \leqslant r_1$, is insignificant. From eq. (2), we get the following expression for $D(r)$:

$$D(r) = \int_0^r 2\pi r\, d(r)\, dr = M(1 - e^{-r/r_0}) \tag{3}$$

When all transport directions are not equally probable—as in the real case discussed below—one can still assume functional forms similar to those of eqs. (1) and (3) for the deposition in each sector measured from the source. We thus define the deposition functions for 45° sectors as

$$d_i(r) = \frac{4 f_i M}{\pi r_{0i}} \frac{1}{r} e^{-r/r_{0i}} \tag{4}$$

and

$$D_i(r) = \int_0^r \frac{2\pi r}{8} d_i(r)\, dr = f_i M(1 - e^{-r/r_{0i}}) \tag{5}$$

The index i refers to the ith sector—in this case

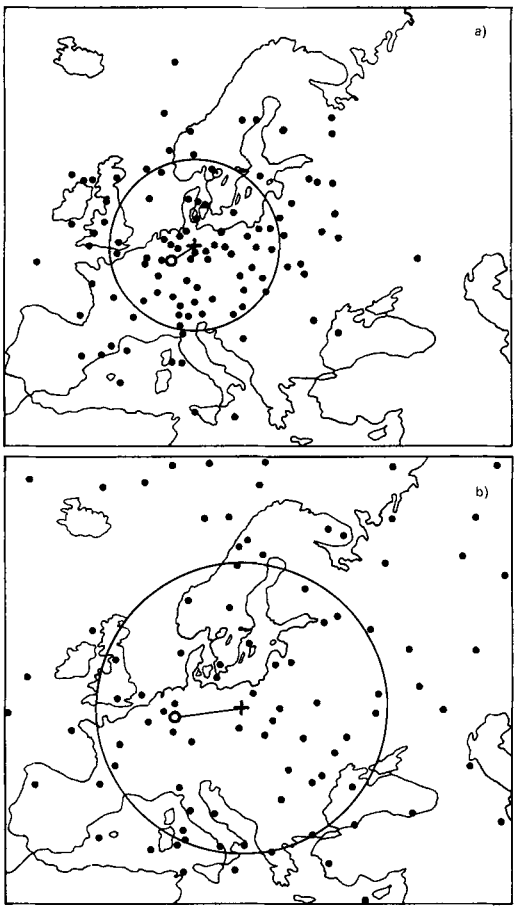

Fig. 4. End points of 850 mb trajectories released from a point (marked by a small circle) in Northern Central Europe for (*a*) 24 hours and (*b*) 60 hours calculated every third day for a period of about one year. 50 % of the points are to be found within the circles which are centered around the mean position of all points (marked with a cross).

45° sectors—and f_i is the frequency of occurrence of trajectory end points (after a certain transport time) in each sector.

To apply eqs. (4) and (5) we thus need to know M, f_i and $r_{0i} = V_i \tau_i$. We have already briefly discussed M in a previous section. The turnover time τ may very well vary between the different transport directions. For example, the probability of rain, can be expected to be higher in the area considered when the winds are from SW or W than when they are from E, thus causing a smaller τ in the former cases. In this study, however, such variations are neglected and in the subsequent applications of equations (4) and (5), τ is assumed to be the same for all transport directions (cf. sections 5 and 6).

Estimates of f_i and V_i can be obtained in the following way. With starting points n four places in Northern Europe (cf. Fig. 5) trajectories have been calculated for approximately every third day during one year. The geostrophic wind approximation has been used and the trajectories have been computed on isobaric levels (850 and 925 mb). Admittedly such trajectories may deviate significantly from the true trajectory of an air parcel which has passed through a city and thus has had an opportunity of carrying away the sulfur which is emitted. Since we shall follow a statistical approach, using many trajectories, we believe that the results are still sufficiently reliable. The largest uncertainty in this connections is probably the uncertainty about the height which should be used for the tra-

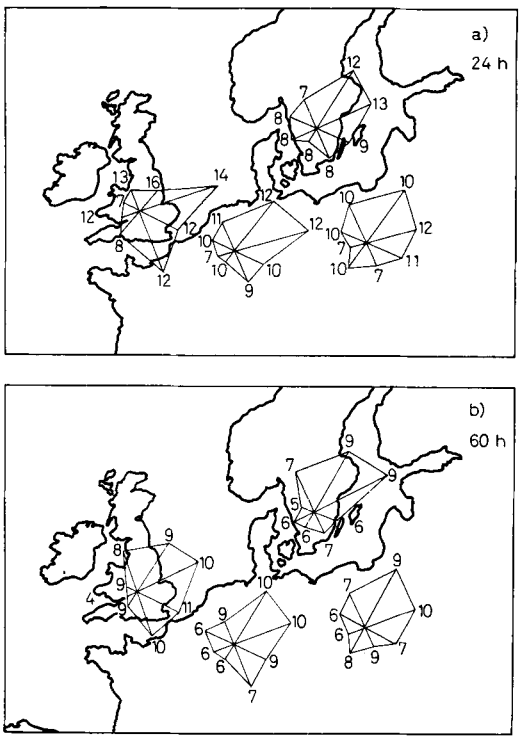

Fig. 5. Frequency distribution (length of lines) of trajectory end points in 45° sectors, and average transport velocities (m/sec) for a travel time of (*a*) 24 hours and (*b*) 60 hours.

jectory computation. Measurements of the vertical distribution of sulfur compounds over more or less polluted areas in Germany (Georgii & Jost, 1964; Georgii, 1970) indicate a rapid decrease with altitude—the SO_2 concentration decreased by a factor of approximately two over the lowest km. Similar measurements over more remote areas in Southern Sweden (Rodhe, 1971) showed a somewhat less distinct decrease with height. It was suggested that, in connection with the study of sulfur transport over long distances, the height chosen for the calculation of trajectories should be around 900 mb (≈ 1 km), if only one height is used.

For this reason both 850 and 925 mb trajectories were computed. It was found that the statistical parameters derived were only slightly different in these two cases. The results from the 850 mb trajectories have been used in the present study.

Fig. 4 shows the distribution of end points after (*a*) 24 hours, and (*b*) 60 hours of such 850 mb trajectories released from a point (marked by a circle) in Northern Central Europe. Similar maps were obtained for trajectories from England, Poland and Southern Sweden. In order to give an over all indication of the mean motion and the rate of dispersion, circles including 50 % of the points centered around the mean position of all points (marked with a cross) are shown.

To calculate f_i and V_i, 45° sectors were defined from each of the starting points. The number of trajectory end points in each sector was counted to give f_i and the average distance from the starting point was measured to give V_i. The results are shown in Fig. 5. The lengths of the lines in the different directions from each point in the figure are proportional to f_i, and the indicated numbers refer to V_i.

5. An estimate of the turnover time for anthropogenic sulfur in the atmosphere

Returning to the sulfur budget for regions I and II, we now want to compare the numbers in Table 2 with the deposition model and try to estimate either the turnover time τ or the length scale r_0.

Let a be the average distance that the anthropogenic sulfur emitted within one of the regions has to travel before it reaches the border of the region. (If, for example, each region had only one source located in the center, a would equal the radius of the circle.) We have used the values 600 km and 1 200 km respectively for a_{I} and a_{II}.

We can now estimate r_0 using the condition that the values of $D(a)$ should equal the percentages in the last column of table 2, which shows the deposition as a percentage of anthropogenic emission for the two regions. The values of r_0 thus obtained are for region I 1 200–2 700 km and for region II 850–2 400 km.

The upper limit for r_0 correspond to the cases when only precipitation deposition is considered. The lower limits are obtained when the total deposition—including dry depositions and gaseous absorption—is assumed to be twice the measured precipitation deposition. The relatively good agreement between the r_0 values for regions I and II may be considered encouraging. With an average transport velocity of 10 m/sec (cf. Fig. 5), the r_0 values correspond to a mean turnover time for anthropogenic sulfur in these regions of 30 to 70 hours.

These values agree reasonably well with the values cited in section 2 from Meetham's and Junge's investigations.

Considerably shorter time scales for the oxidation rate of SO_2 have been observed or derived by other investigators (e.g. Gartrell et al., 1963; Weber, 1970). When comparing these figures it must be kept in mind that these short time scales refer mainly to the transformation of SO_2 into sulfate; the total life time for sulfur is certainly longer. Furthermore, it is conceivable that under some conditions—especially at high relative humidity and when there are high concentrations of other pollutants in the immediate vicinity of a sulfur source—the life time for some of the emitted sulfur is short while the effective mean value is still of the order of a few days.

Rodhe et al. (1971) compared sulfate concentrations in the air of some Swedish cities with corresponding values from remote locations in the same area. They found that on the average, only about 5% of the emitted SO_2 had been oxidized and transformed into sulfate aerosols while the air remained in the city, i.e. within the first 10–20 min after emission.

Table 4. *Anthropogenic sulfur emission data for some regions in Northern Europe, used for the estimation of the sulfur deposition in Sweden. (Unit: 10^6 tons S/year)*

A	U.K. and Ireland	3.2
B	North-Western Central Europe	3.0
C	North-Eastern Central Europe	1.5
D	Southern Sweden (south of lat. 61° N)	0.25
E	Northern Sweden	0.1

6. The contribution from different sources to the sulfur deposition over Sweden

As an illustrative example, some of the information derived in the previous sections will be used to make a rough estimate of the relative importance of different sources for the deposition of sulfur over Sweden.

Since there is a very marked gradient in sulfur deposition in the north–south direction over Sweden, two separate regions were considered. The common boundary for the two regions was the line of latitude 61°N. Table 4 shows the values for the anthropogenic sulfur emission which were used (cf. Tables 2 and 3).

When calculating the sulfur deposition in Southern and Northern Sweden respectively eq. (4) and (5) were used and all anthropogenic emissions were assumed to originate from five point sources, located as in Fig. 5 and with the source strengths given in Table 4. The values for f_i were obtained from Fig. 5 and, in accordance with the results of the previous section, two values for r_0 were chosen, namely 1 000 and 2 600 km. The results of the calculations are shown in Table 5.

It is recalled that the background values refer to natural sources and to anthropogenic sources outside Northern Europe.

It is of interest to compare the totals in Table 5 with the observed sulfur deposition. From Fig. 3 it may be estimated that the precipitation deposition in the middle of the sixties was about 950 and 550 mg/m² per year for Southern and Northern Sweden respectively. Since r_0 = 1 000 km was derived on the assumption that the total deposition is twice as large as that through precipitation deposition only, the totals in the first two columns of table 5 should be compared with the amounts estimated on the basis of observations, i.e. 1 900 and 1 100 mg/m² per year. The use of r_0 = 1 000 km yields calculated deposition values which are significantly lower than the deposition estimated on the basis of observations. On the other hand, it is seen that the deposition model gives a roughly consistent picture for r_0 = 2 600 km.

We conclude that the most probable value for r_0 is about 2 500 km (corresponding to a turnover time of some 70 hours). With this value for r_0 the deposition budget shown in Table 6 is obtained.

Admittedly, the uncertainties in the derived numbers are substantial, not least because of the uncertainty in the assumption about the background deposition. If the background deposition is larger than 250 mg/m² per year,

Table 5. *Estimated contributions from different sources to the sulfur deposition in Southern and Northern Sweden for two different values of the deposition length scale r_0 (around the year 1965). (Unit: mg S/m² per year)*

	r_0 = 1 000 km		r_0 = 2 600 km	
Source	S Sweden	N Sweden	S Sweden	N Sweden
U.K. and Ireland	220	70	180	80
NW Centr. Europe	290	90	210	90
NE Centr. Europe	120	50	80	50
S Sweden	290	50	120	30
N Sweden	10	100	5	40
Background	250	250	250	250
Total	1 180	610	845	540

Table 6. *Estimated contributions from different sources to the sulfur deposition in Southern and Northern Sweden around the year 1965. The deposition length scale (see text) have been assumed to be 2 500 km. (Unit: mg S/m² per year)*

Source	S Sweden	N Sweden
Foreign emissions	480	230
Swedish emissions	140	80
Background	250	250
Total	870	560

Table 7. *Estimate of the uncertainty of the values given in Table 6. The numbers have been normalized with respect to the total deposition*

Source	S Sweden	N Sweden
Foreign emissions	0.32–0.63	0.30–0.45
Swedish emissions	0.08–0.22	0.07–0.19
Background	0.60–0.15	0.63–0.37
Total	1	1

in this part of the world, it would not only affect the background values themselves but also have the effect of increasing the estimate of r_0, thereby decreasing the other values in Tables 5 and 6. The assumption concerning the specific form of the deposition function is believed to be less critical for the results obtained. Slightly different forms have been tried, yielding different r_0-values, but roughly the same deposition budget has resulted from each.

An estimated range of uncertainty is shown in Table 7, where the values have been normalized with the total deposition as unity. This estimate has been based on a probable interval for r_0 of 1 500–5 000 km.

It may seem that there is a contradiction between the results shown in Table 3, which indicate that the anthropogenic sulfur emissions in Southern Sweden seem to be larger than the total deposition within the same area and the results shown in Table 6. However, this is explained by the fact that a considerable part of the sulfur emitted in Sweden is exported out of the country before it is deposited.

It may be worth emphasizing that there is no one to one correspondence between the amount of sulfur deposited on the ground and the acidity of precipitation that may be harmful to terrestrial and aquatic ecosystems. Other chemical compounds in air and rainwater are also important. Therefore, where deposition of acid is concerned, the distribution between different sources may be different from that shown for sulfur in Table 6.

It is furthermore important to realize that we have studied only the integrated deposition over Southern and Northern Sweden. The magnitudes of the effects of an increased deposition locally, in the neighbourhood of strong Swedish sulfur sources, has not been discussed. Inclusion of this effect may alter to some extent the distribution of "responsibility" as it is shown in Table 6.

7. Conclusions

The present study of the atmospheric sulfur budget for a region with large anthropogenic emissions shows clearly that the distribution of sulfur from these sources has a regional character, and that strong anthropogenic sulfur sources can thus contribute significantly to the amount of deposition thousands of kms away. This does not mean that harmful effects of high concentrations of SO_2 *in the air* occur at great distances from a source—the rapid decrease in concentration that takes place by turbulent mixing is normally (in not too densly industrialized areas) sufficient to confine possible damage of this kind to the close surroundings of the source (a few kms).

In the case of possible harmful effects caused by the acidity of precipitation the situation is however quite different. The deposition of sulfur, which, if the sulfuric acid is not neutralized by alkaline compounds, may be associated with a deposition of acid, is not so markedly concentrated in the close surroundings of a source, but is distributed more uniformly over large areas. Because of the cumulative effects the deposition of acid can give rise to adverse effects even far away from anthropogenic sources.

The reason why the deposition of sulfur and acid does not show such a pronounced maximum in the closest surrounding of a sulfur source as does the concentration of sulfur dioxide, seem to be the following:

(1) The sulfur that is deposited through precipitation originates, to a large extent, from the rain-producing layers (1–5 km) where the concentration is much less influenced by local emissions than is the surface air. (See e.g. Bielke & Georgii, 1968.)

(2) A large part of the SO_2 is probably not deposited until after it has become oxidized to form sulfate aerosols. The rate of oxidation is very variable and not well known, but measurements indicate that sulfate aerosols are created gradually in the air and that the concentration of such aerosols is not very much higher near the sources than in the remote areas

within a given region (Georgii, 1970; Rodhe et al., 1971).

In this study the budget calculations for two regions in Northern Europe indicate that there must be a considerable net outflow of anthropogenic sulfur through the circle of about 1 000 km radius which encloses the large anthropogenic sulfur sources in U.K. and Northren Central Europe.

A model for the variation of the deposition with distance from a source has been discussed. This model, when applied to some areas in Northern Europe for which sulfur budget calculations have been made, gives a length scale for the rate of decrease of deposition with distance from the large anthropogenic sources of 2 000–3 500 kms. With a typical horizontal transport velocity of 10 m/s at a height of about 1 km this corresponds to a turnover time for anthropogenic sulfur of 2 to 4 days.

In order to illustrate the effect of the long range transport of sulfur, an estimate has been made of how different anthropogenic sulfur sources contribute to the deposition over Sweden. This calculation has been based on an estimated turnover time of 3 days (cf. above) and on statistics regarding the routes of transports (trajectories) in this region.

The results indicate that especially for Southern Sweden the size of the sulfur deposition is significantly affected by foreign anthropogenic sources; the relation being approximately 30/20/50 between the contributions from natural, Swedish anthropogenic and foreign anthropogenic sources respectively.

Just as Sweden is thus affected from foreign sulfur emissions, a large part of the Swedish emissions are deposited outside the country.

8. Acknowledgements

This study has been undertaken in order to give background material to a case study which has been prepared by a group of Swedish scientists for the UN conference on the human environment 1972. I appreciate helpful discussions with all members of this group, especially Professor Bert Bolin, Mr Lennart Granat and professor Svante Odén. Dr Peter Davies has been kind enough to assist in improving the language.

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ИЗУЧЕНЕ ИБАЛАНСА СОДЕРЖАНИЯ СЕРЫ В АТМОСФЕРЕ НАД СЕВЕРНОЙ ЕВРОПОЙ

Обсуждается баланс содержания серы в атмосфере над Северной Европой. Концентрации серы сравниваются с глобальными средними значениями. Найдено, что в этом районе источники серы антропогенного происхождения превалируют над естественными источниками. Сравнение между отложением и выделением серы в различных частях Северной Европы показывает, что рассеяние серы имеет континентальный характер, т. е. поскольку в среднем сера переносится на расстояния более 1 000 км, прежде чем она осаждается, то в этом отношении одна страна может влиять на другую. Предложена предварительная модель для изменения отложения с расстоянием от источника. Пользуясь этой моделью можно получить оценку времени обращения антропогенной серы в 2–4 дня. В виде качественного примера для Швеции оценено соотношение между различными источниками отложений серы. Расчеты показывают, что около половины этой серы выделяется иностранными антропогенными источниками, в то время как другая половина выделяется шведскими и естественными источниками. Точно также, как Швеция подвергается влиянию окружающих ее индустриальных стран, так и большая часть серы, выделенной в Швеции, осаждается вне страны.