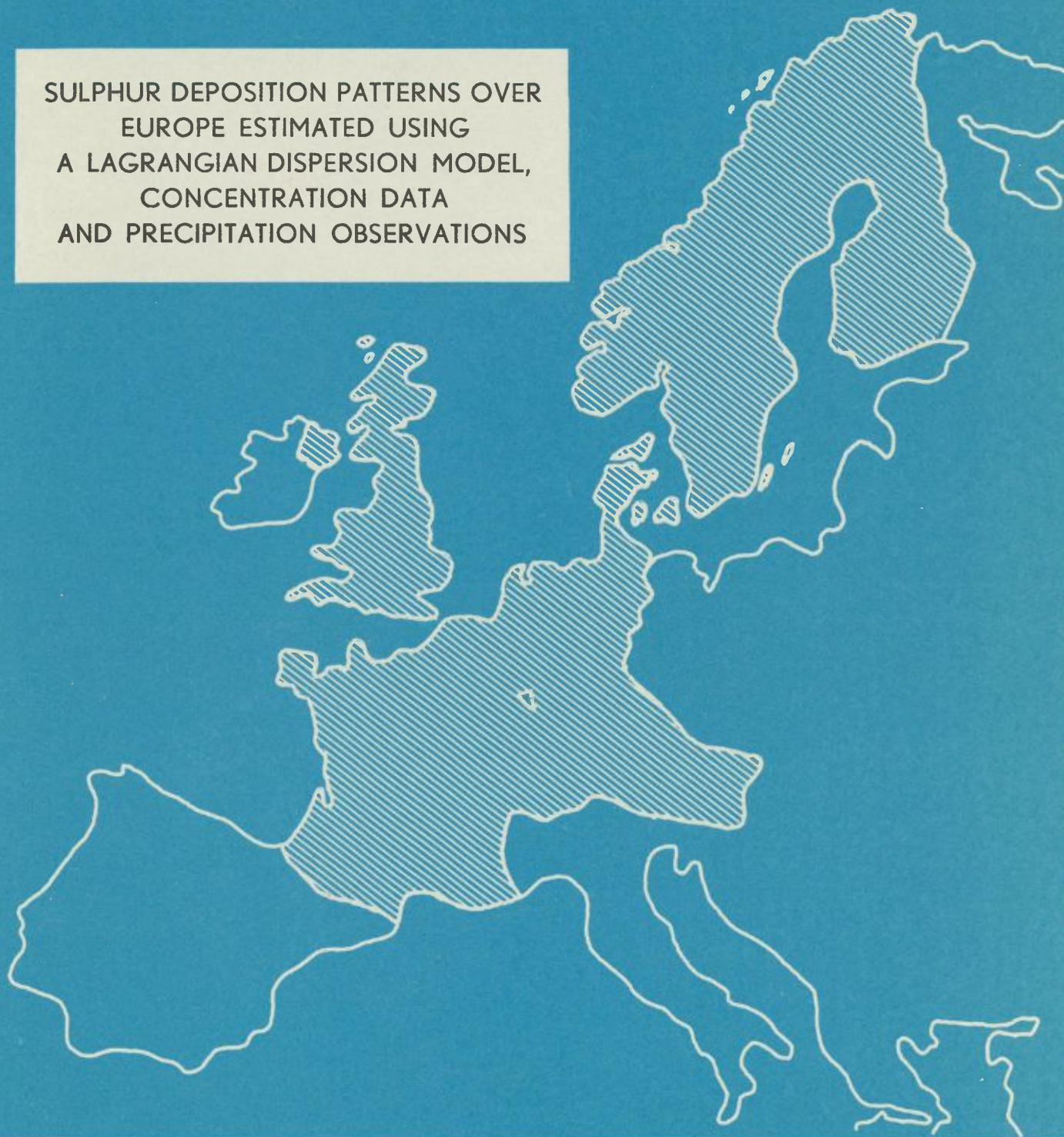# LONG RANGE TRANSPORT OF AIR POLLUTANTS

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SULPHUR DEPOSITION PATTERNS OVER EUROPE ESTIMATED USING A LAGRANGIAN DISPERSION MODEL, CONCENTRATION DATA AND PRECIPITATION OBSERVATIONS

SULPHUR DEPOSITION PATTERNS OVER EUROPE ESTIMATED USING A LAGRANGIAN DISPERSION MODEL, CONCENTRATION DATA AND PRECIPITATION OBSERVATIONS

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SULPHUR DEPOSITION PATTERNS OVER
EUROPE ESTIMATED USING A LAGRANGIAN
DISPERSION MODEL, CONCENTRATION DATA AND
PRECIPITATION OBSERVATIONS

1 INTRODUCTION

In the following an estimated sulphur wet deposition pattern
for Europe is presented, covering the period 13th December
1973 to 1st April 1975. The wet deposition pattern is based
on precipitation data from the meteorological stations net-
work, calculated SO_2 air concentrations from a Lagrangian
dispersion model, and an empirical relationship established
between mean calculated SO_2-concentrations in air and mean
measured sulphate concentrations in precipitation.

Using the Lagrangian dispersion model, a SO_2 dry deposition
pattern has been calculated for the same period. Half-year
SO_2 dry deposition patterns for the period July 1972 - July
1975 are presented elsewhere (Eliassen and Saltbones 1975),
together with a discussion of the model and some other
results.

The total amounts of sulphur emitted and deposited within
the area considered during the period have been estimated
using SO_2 emission data for Europe and the two calculated
deposition patterns.

2 THE SULPHATE WET DEPOSITION PATTERN

2.1 Calculation

The total sulphate wet deposition S per unit area at any
place within the region considered is:

$$S = \sum_i p_i c_i = P\bar{c} \tag{1}$$

where p_i is the amount of precipitation fallen and c_i is
the concentration of sulphate in precipitation at day no. i,
$\bar{c}$ is the mean (weighted) sulphate concentration and P is the
total amount of precipitation fallen during the period. The
quantities p_i and P may be evaluated using data from the net-
work of meteorological stations. This network covers Europe
fairly well. Every day observations of precipitation from
500-1000 stations have been employed to construct daily pre-
cipitation fields in a 127 km grid. A method of objective
analysis has been employed, with a "distance of influence"
of 300 km over the sea and 100 km over land. Over the sea,
the precipitation probably is underestimated in some areas
due to lack of data. Figure 1 shows the field of total
precipitation P during the period, obtained by adding all
the daily precipitation fields.

Values of both p_i and c_i throughout the period are available
at 50 LRTAP sampling sites. To calculate the wet deposition
of sulphate in the large areas not covered by these sites,
estimated values of sulphate concentrations in precipitation
are needed. For this purpose, some relation is sought between
the mean sulphate concentration $\bar{c}$ and calculated air concen-
trations from the Lagrangian dispersion model. At 26 of the
LRTAP sampling sites, calculated daily SO_2 air concentrations
q_i from the Lagrangian dispersion model are available together
with p_i and c_i. Figure 2 shows the yearly mean 1973 concen-
trations $\bar{c}$ at these stations plotted against the yearly weighted
mean SO_2-concentrations $\bar{q}$, defined by:

$$P\bar{q} = \sum_i p_i q_i \qquad (2)$$

Values of $\bar{c}$, $\bar{q}$ and P at the various sites for 1973 and 1974
are given in Table 1. The measurements of sulphate concen-
trations in precipitation at Danish stations are not con-
sidered reliable during the second half of 1974. From Table 1
it is seen that $\bar{c}$ has increased by large amounts from 1973
to 1974 at the Danish sites. A corresponding increase

has not taken place at other sites. This increase in mean
values at the Danish sites is accompanied by a significant
increase in standard deviations.

From Figure 2 it is seen that there is a fairly good relation-
ship between $\bar{c}$ and $\bar{q}$. The relationship is obviously not
linear, and a curve is drawn by hand to fit the data.

It should be mentioned that the Lagrangian model used to
calculate the SO_2 air concentrations q_i does not include
any specific description of the removal of SO_2 by precipitation.
In the model calculations, SO_2 is assumed to have a constant
half-life of 19 hours.

Since p_i and q_i are available for all grid elements in the
127 km grid, a value of $\bar{q}$ may be calculated for each ele-
ment, and an estimate of the mean sulphate concentration $\bar{c}$
may be obtained by assuming that the empirical relationship
of Figure 2 is valid everywhere. The total sulphate wet
deposition S at the grid element is found by multiplication
with the total amount P of precipitation. The resulting
sulphate wet deposition pattern is shown on Figure 3. The
measured sulphate wet deposition at LRTAP sites are shown
on Figure 4.

2.2 Discussion

The sulphate wet deposition pattern of Figure 3 shows maxima
in the Ruhr region, in Southern Germany, in Southern
Scotland and in Southern Norway. Although the wet deposition
pattern largely resembles the precipitation distribution
of Figure 1, some differences are clearly visible. In Northern
Norway, North-Western Scotland and Ireland, large values of
P are not reflected by large values of sulphate wet deposition.
The reason for this is obviously that the estimated sulphate
concentration in rain is low in these areas. For the same
reason, the maximum zone of precipitation in Southern Norway

is found along the west coast, whereas the maximum sulphate
wet deposition is found on the southeastern slope of the
mountains. Here, the estimated sulphate concentration in the
rain is higher, because on rainy days the air has usually
passed some of the large SO_2-emission areas in western or
central Europe.

Assuming that the measured depositions of Figure 4 are
representative for the surrounding regions, it is seen that
the estimated deposition level of Figure 3 is somewhat high
in Great Britain and somewhat low in France, but that the
estimated and measured depositions may reflect the same geo-
graphical distribution. In the other regions, there is a good
agreement between estimated and measured depositions.

Calculated patterns of sulphur wet deposition have been
presented earlier by Bolin and Persson (1973). They used a
statistical formulation of the transport equation, and cal-
culated the horizontal dispersion of sulphur on the basis
of the statistical properties of a large number of trajectories
initiated from different points in Europe. As a first
approximation, the processes of dry and wet deposition of
sulphur were described using constant deposition rates.

The main difference between the wet deposition patterns of
Bolin and Persson, and the patterns presented here, is that
the wet deposition maximum in Southern Norway does not
appear in their calculations. There are two main reasons for
the existence of this maximum: a) When a low pressure cell,
to which precipitation is associated, approaches Southern
Norway from the west, a southerly airstream is set up flowing
from areas with large SO_2-emissions towards Scandinavia,
b) When winds are southerly, there is a high probability for
precipitation to occur in Southern Norway due to orographic
effects. These effects cannot be described by a wet deposition
rate independent of position and wind direction.

3   BUDGET CONSIDERATIONS

The SO_2 emission field in this work is shown on Figure 5. This emission field is mainly an extension of the field employed earlier by Eliassen and Saltbones (1975b) to a larger area. The yearly emission data are believed to be within ±20% of the actual figures, but may be more uncertain for the eastern european countries. Natural sources of SO_2 have not been taken into account. In the region considered they are probably negligible compared to the anthropogenic ones.

The SO_2 dry deposition pattern for the period is shown on Figure 6. Mean SO_2-concentrations are calculated using the Lagrangian dispersion model mentioned earlier, and a constant deposition velocity of 8 mms^{-1} is employed. A description of the dispersion model, and dry deposition maps for other periods are presented elsewhere (Eliassen and Saltbones 1975). It should be mentioned, however, that the dry deposition values may be too small in the major emission areas. When calculating the dry deposition, the computed mean concentrations are adjusted by comparing with measured mean concentrations at LRTAP sampling sites. To avoid that the influence of long range transport is masked by contributions from local emissions, these sites are placed in rural areas and may therefore not be representative for the ground level concentration of a grid element. The importance of this effect depends upon the distribution of the ground level concentration inside the grid elements. Information on such distributions are not readily available, thus the evaluation of this effect would require extensive additional investigations on the sub-grid scale, taking into consideration that a significant part of the emissions are warm emissions from tall stacks, and that urban plumes may be lifted from the surface by thermal effects.

Figure 7 shows the total sulphur deposition pattern, obtained
by adding the dry deposition of SO_2 and the wet deposition
of sulphate. The dry deposition of particulate sulphate is
probably less than 10% of the SO_2 dry deposition in the region
considered, and are neglected in the budget considerations.
The reason for this is that the deposition velocity of sul-
phate particles, which have diameters typically in the
0.1 - 1 μm range, is an order of magnitude lower than the
value of about 1 cms^{-1} established for SO_2 (Chamberlain, 1966),
(Owers and Powell, 1975), (Shepherd, 1975), (Garland et al.
1975), and the measured particulate sulphate concentrations
in air is generally lower than the measured SO_2-concentra-
tions.

From the emission field, the total emission of SO_2 during
the period (13.12.73 to 1.4.75, 474 days) is $6.8 \cdot 10^7$ metric
tonnes. The total dry deposition of SO_2 is $3.0 \cdot 10^7$ tonnes,
or 44% of the emitted amount. The total wet deposition of
sulphate is $1.5 \cdot 10^7$ tonnes (22%). The total flux of sulphur
out of the region is $2.3 \cdot 10^7$ tonnes (34%), estimated as a
difference between total emissions and total deposition.
(All numbers in tonnes of SO_2.)

Figure 8 shows the difference between the total deposition
and the emission of sulphur for each grid element during
the period. Positive numbers signify a net import of sul-
phur, and negative numbers a net export. The main export
regions are confined to the industrialized regions of
Central Europe and Great Britain.

Table 2 shows the total emission and estimated total depo-
sition for each country. As one should expect, most countries
in western Europe are net exporters of sulphur. According to
these estimates, Norway imports a large amount of sulphur
compared to its own emissions.

4   CONCLUSIONS

Even though the sulphur deposition patterns presented here
are valid for the period 13.12.73 to 1.4.75, they show
some traits which may be more generally valid when con-
sidering time periods of a year or more. Within the
region considered, the dry deposition of SO_2 is about twice
as large as the wet deposition of sulphate (in SO_2 mass units).
In regions with large emissions of SO_2, dry deposition of
SO_2 is the most important deposition process. Some regions
with small emissions, however, are located in such a way
that when precipitation is released, there is a high
probability that the air masses have passed over areas
with large emissions of SO_2. In such regions, the wet depo-
sition of sulphate may outweigh the dry deposition of SO_2.

Typical examples are Southern Norway, and to some extent
Southern Scotland, and the northward and southward slopes
of the Alps. In Southern Norway, the wet deposition is three
times as high as the dry deposition, and in the other
regions mentioned the wet and dry depositions are of about
the same magnitude.

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Western Europe".
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Caughey, S.J. (1974)
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Owers, S.J. and Powell, A.W. (1974)
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surfaces using a ^{35}S tracer method".
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Shepherd, J.G. (1974)
"Measurements of the direct deposition of sulphur dioxide
onto grass and water surfaces by the profile method".
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	$\bar{c}$ $(\mathrm{mg}\,\ell^{-1})$		$\bar{q}$ $(\mu\mathrm{gm}^{-3})$		$P\,(\mathrm{mm})$	
	1973	1974	1973	1974	1973	1974
A 1	14.7	–	41.9	–	104	–
A 2	7.3	10.2	38.4	38.0	127	103
D 3	–	3.6	–	18.0	–	170
DK2	4.2	9.4	8.7	10.8	423	706
DK3	3.4	15.2	10.9	12.1	480	613
DK4	3.7	8.9	16.7	19.8	390	552
DK5	4.5	7.0	19.0	16.9	264	402
DK6	5.8	9.1	19.3	17.1	381	497
F 1	5.5	5.7	15.3	17.4	529	650
N 1	3.3	3.4	11.5	12.1	1030	1550
N 3	2.3	2.6	9.1	10.4	1160	1640
N 9	1.7	1.6	3.7	4.8	2020	1780
N22	3.9	3.4	10.3	12.1	493	682
N23	3.9	4.0	12.1	11.6	678	808
NL1	6.0	5.8	29.6	27.3	601	788
NL2	3.5	4.2	23.5	25.8	733	810
NL3	4.9	3.8	20.7	20.0	507	796
NL4	–	5.6	–	25.9	–	727
SF1	3.0	2.9	7.9	11.1	560	766
SF2	3.5	2.8	10.5	11.0	533	614
SF3	2.8	3.8	8.1	11.3	522	849
SF4	2.7	2.0	6.6	8.4	613	701
SF5	1.5	1.5	2.7	3.4	344	541
S 3	3.7	4.2	11.0	14.9	469	689
S 4	3.3	3.4	9.3	11.3	473	578
S 5	1.5	1.9	3.5	4.8	669	876
UK1	4.8	3.7	38.3	36.3	440	688
UK2	2.6	1.9	10.3	9.8	792	1120

Table 1: Mean measured sulphate concentrations in precipitation $\bar{c}$, mean calculated SO_2-concentrations $\bar{q}$ defined by eq. (2), and total amount of precipitation at some LRTAP sites for the years 1973 and 1974. The sulphate concentrations have been corrected for sea spray where this is considered necessary.

The station A 1 in Austria was discontinued in August 1973, and was succeeded by another station in the same region (A 2).

COUNTRY	EMISSION 10³T SO₂	DRY DEPOSITION 10³T SO₂	WET DEPOSITION 10³T SO₂	TOTAL DEPOSITION 10³T SO₂	TOTAL DEPOSITION EMISSION %
AUSTRIA	430	470	300	770	180
BELGIUM	1400	260	130	390	30
DENMARK	650	150	90	240	35
FINLAND	630	410	540	950	150
FRANCE	3900	1500	800	2300	60
WESTERN GERMANY	5400	2000	1000	3000	55
ITALY	4200	1100	400	1500	35
THE NETHERLANDS	1200	270	130	400	35
NORWAY	230	250	490	740	320
SWEDEN	1300	720	780	2500	120
SWITZERLAND	190	180	130	310	160
UNITED KINGDOM	8600	1100	700	1800	20

Table 2: Estimated emissions and depositions of sulphur for each of the countries, for the period 13.12.73 to 1.4.75 (1.3 years). The uncertainties are believed to be around 20% for the emitted amounts, and probably larger for the estimated depositions.
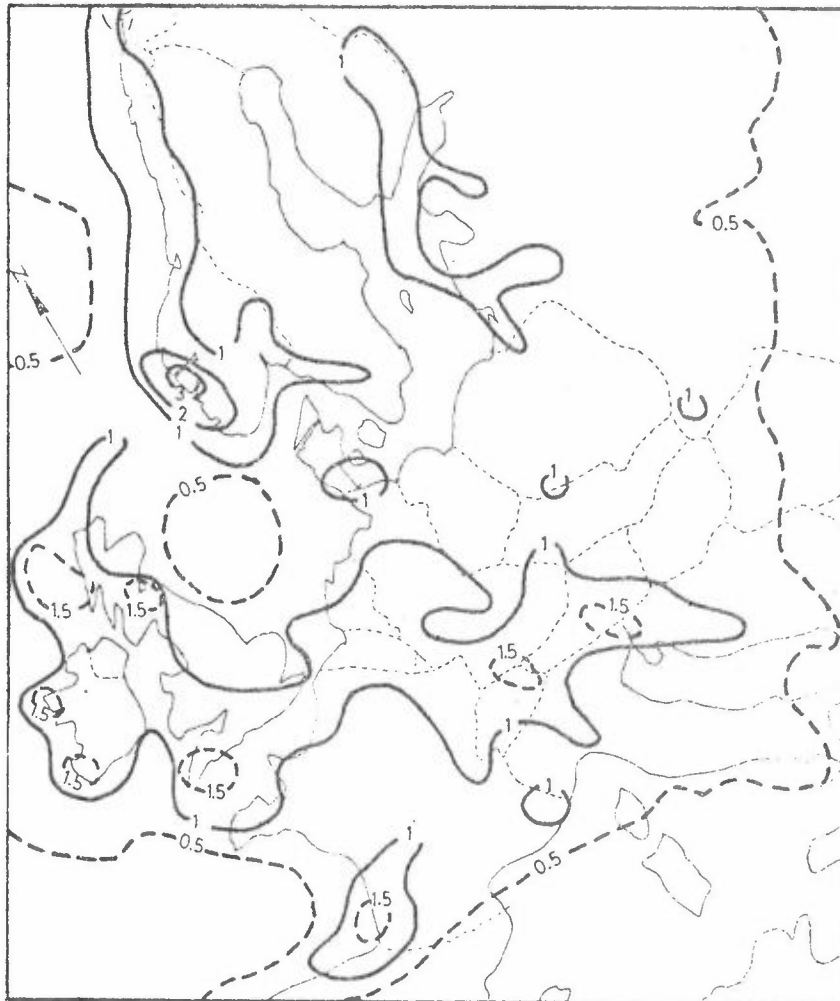
Figure 1: Total precipitation P(m) for the
period 13.12.73 to 1.4.75, obtained
by adding all the daily precipitation
fields. The values over the sea result
from the method of objective analysis
employed, with a "distance of in-
fluence" for precipitation data of
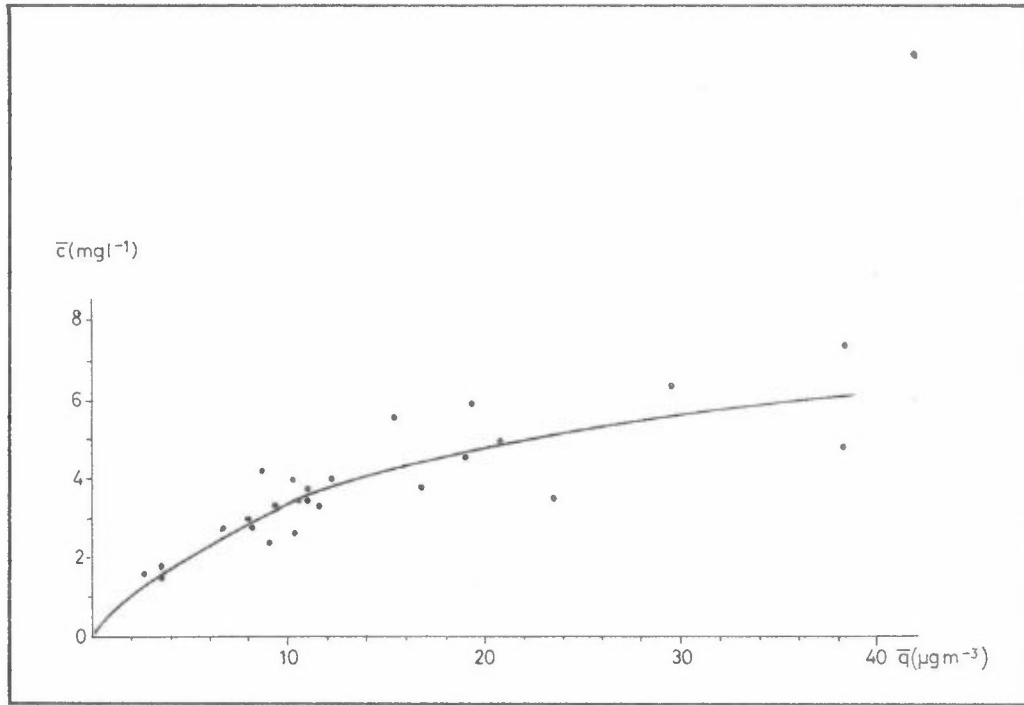300 km over the sea, and 100 km over
land.

Figure 2: Mean observed sulphate concentrations
$\bar{c}$ in precipitation plotted against mean
computed SO_2-concentrations $\bar{q}$ at some
LRTAP sampling sites using 1973-data.
Both means are weighted, using daily
precipitation measurements (see text).
Values of $\bar{c}$ and $\bar{q}$ for 1973 and 1974
are given in Table 1.
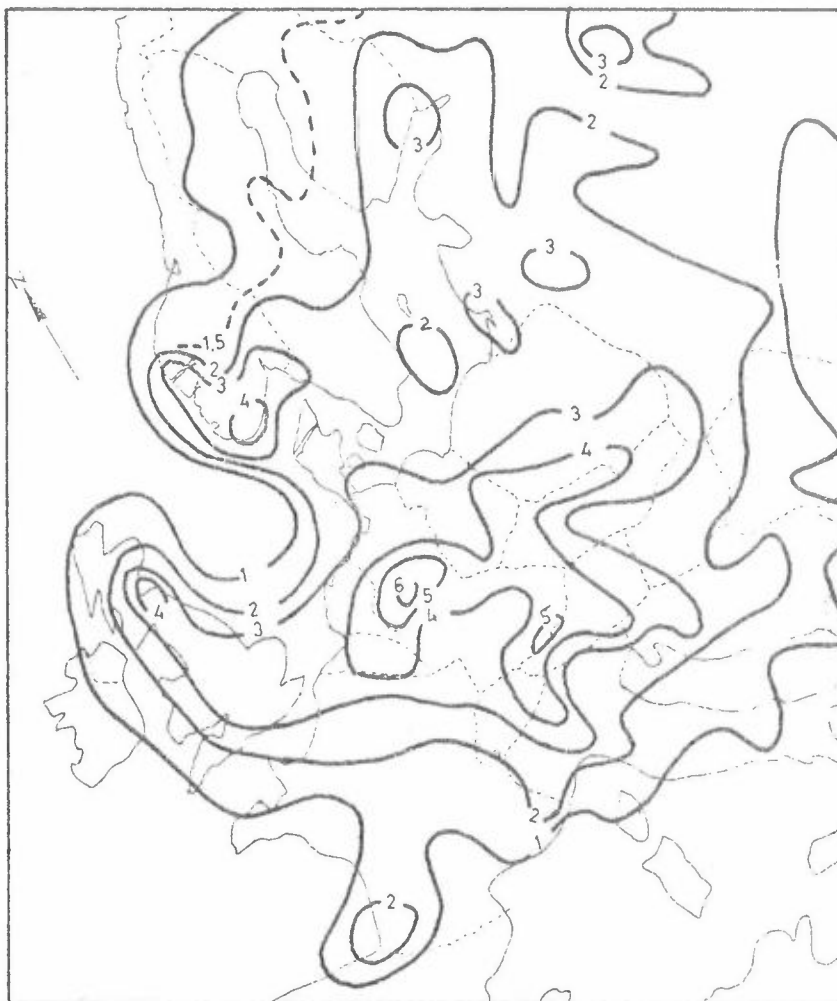
Figure 3: Estimated sulphate wet deposition
pattern for the period 13.12.73 to
1.4.75. Unit: g SO_2 m^{-2}. The
minimum in the North Sea arises
from the lack of precipitation data
over the sea, see Figure 1 and text.
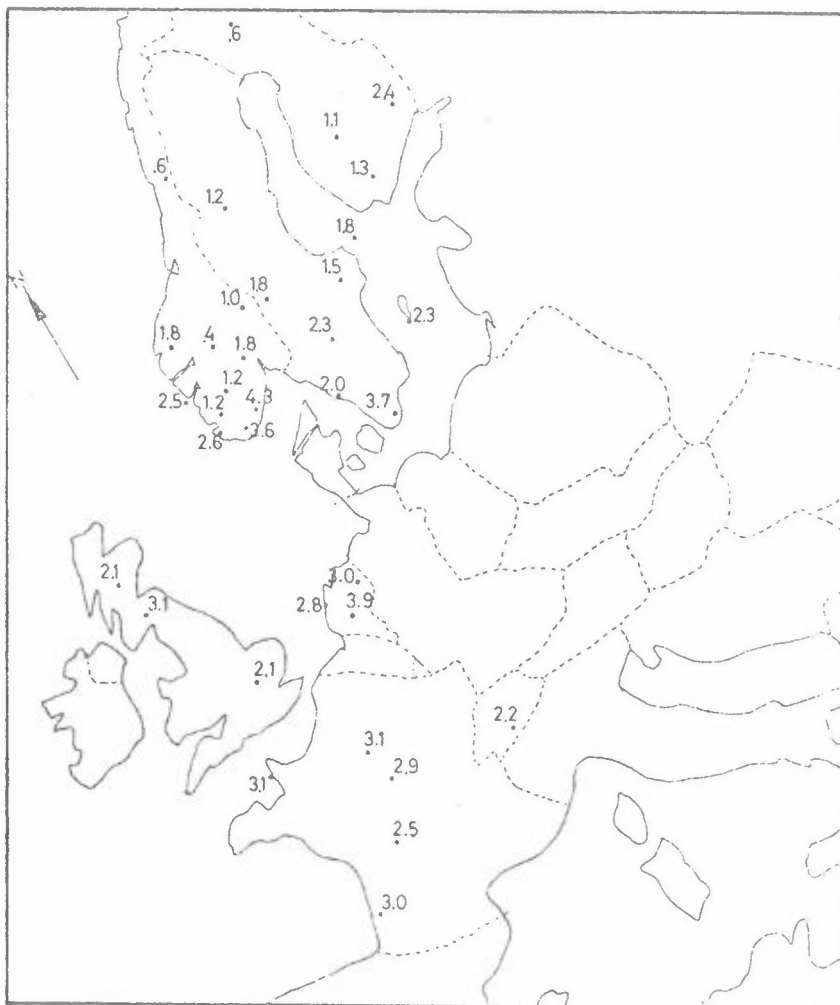
Figure 4: Measured sulphate wet deposition at
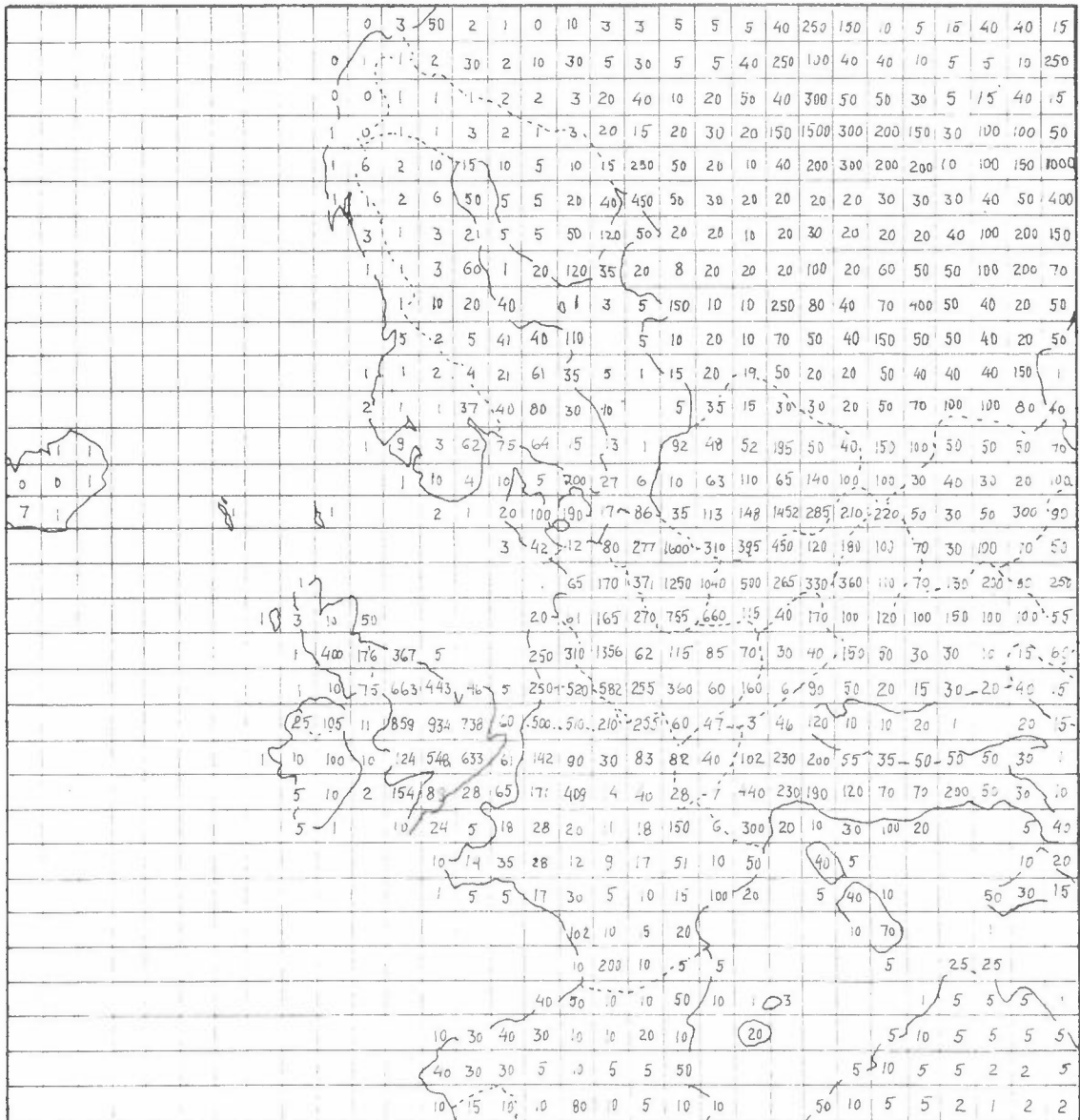LRTAP sampling sites, same period as
Figure 3.
Unit: g $SO_2 m^{-2}$.

Figure 5: SO_2-emissions used in this work. The
data are believed to be within ±20%
of the actual figures, but may be more
uncertain for the eastern european
countries. Natural sources have not
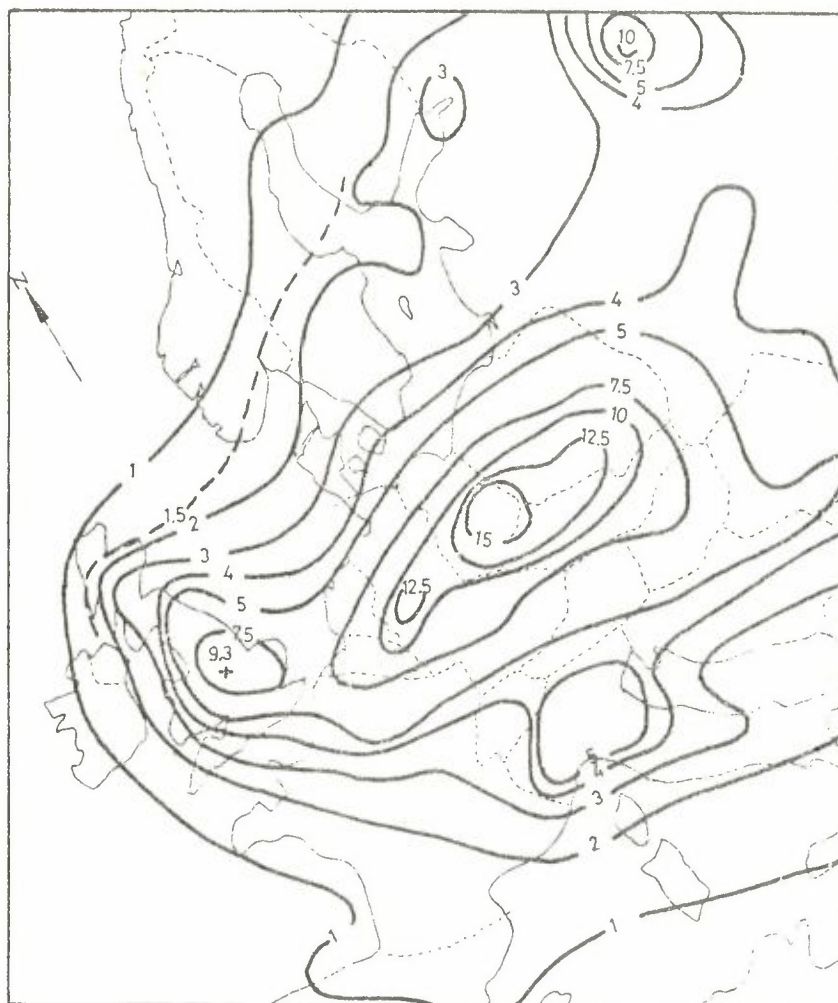been taken into account.
Unit: 10^3 tonnes $SO_2 yr^{-1}$.

Figure 6: Estimated SO_2 dry deposition pattern for
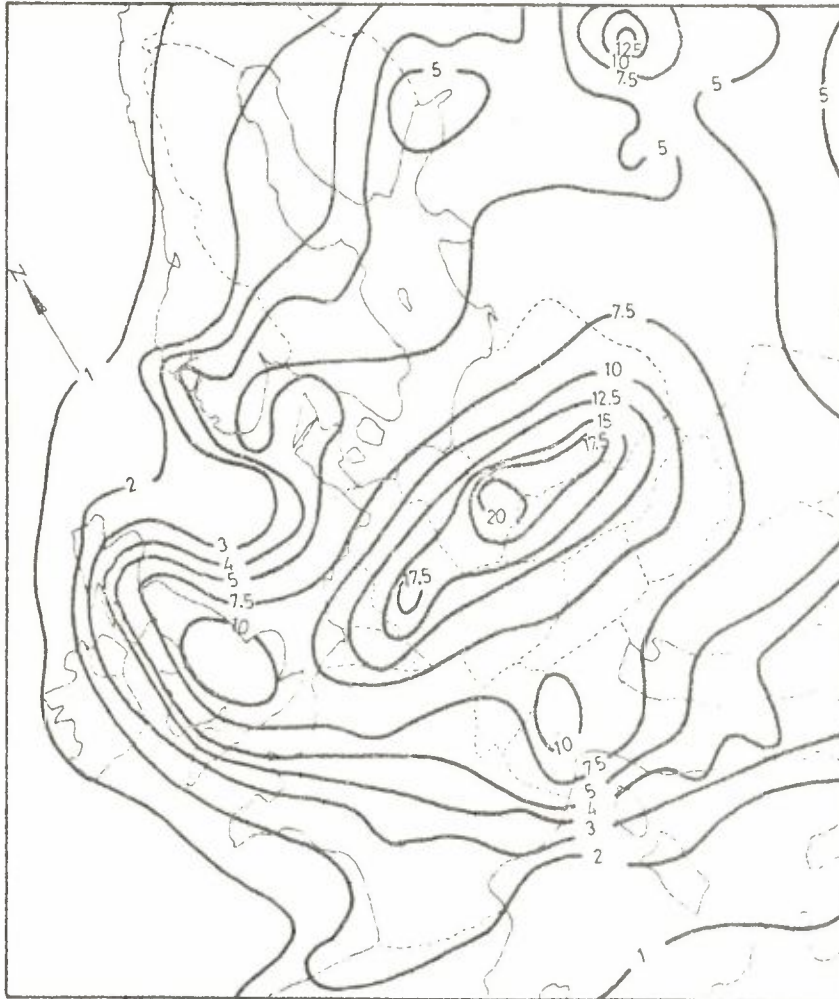the period 13.12.73 to 1.4.75.
Unit: g $SO_2 m^{-2}$.

Figure 7: Total sulphur deposition pattern, obtained by adding the wet deposition of Figure 3 and the dry deposition of Figure 6. Dry deposition of particulate sulphate is neglected. Unit: g $SO_2 m^{-2}$.
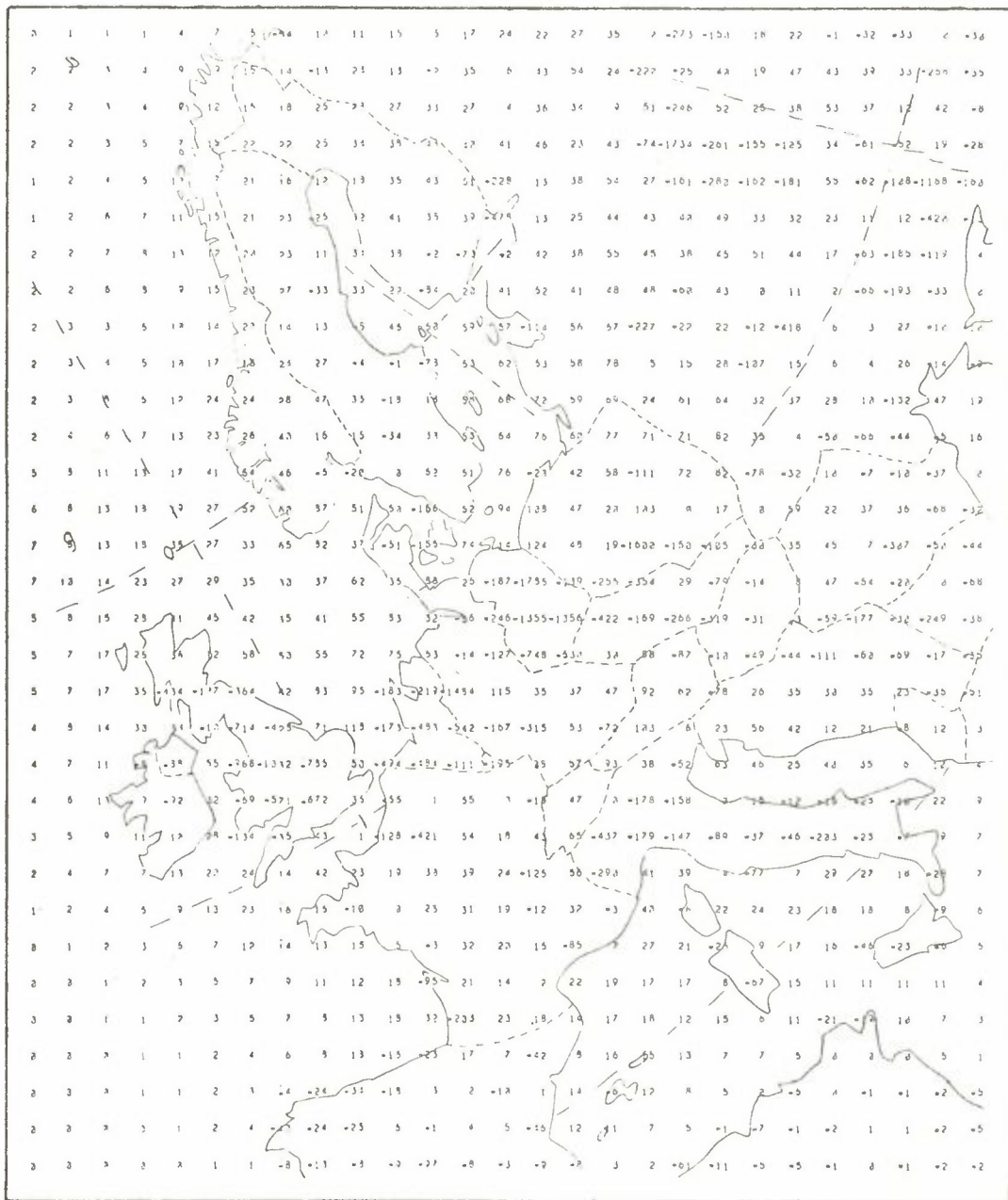
Figure 8: Difference between total deposition and emission
of sulphur for each grid element during the
period. Unit: 10^3 tonnes SO_2.