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A SIMPLE LAGRANGIAN DISPERSION MODEL  
APPLIED TO SULPHUR POLLUTION OVER EUROPE

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ABSTRACT

A simple Lagrangian dispersion model is described and applied to sulphur pollution over Europe. The model calculations are based on available SO<sub>2</sub>-emission data for Europe, and wind observations in the 850 mb surface. A special case is reported where the presence of computed large-scale SO<sub>2</sub> and SO<sub>4</sub> plumes are verified by concentration data from aircraft sampling and from the OECD sampling network. For stations in this network, computed and observed daily mean SO<sub>2</sub> and SO<sub>4</sub> concentrations are compared for a period of six months. Based on this comparison an SO<sub>2</sub> dry deposition pattern for Europe for 1974 is calculated, using computed mean concentrations and a deposition velocity of 0.8 cms<sup>-1</sup>.

## INTRODUCTION

As part of the OECD-project "Long Range Transport of Air Pollutants" (LRTAP) a network of sampling stations have been set up in the participating countries. The data obtained from this network are daily mean concentrations of chemical components in precipitation and air. Two components in air are measured: SO<sub>2</sub> and particulate SO<sub>4</sub>. Surveys of anthropogenic SO<sub>2</sub>-emission within Europe have been carried out in connection with the project. Atmospheric dispersion models have been developed to link the emission surveys and the observed concentrations.

In the following a simple Lagrangian-type dispersion model is described. The model includes a transformation SO<sub>2</sub> → SO<sub>4</sub> and thus gives estimates of SO<sub>2</sub> and SO<sub>4</sub> air concentrations. As an example the model calculations are compared with observed concentrations in a situation with large-scale SO<sub>2</sub> and SO<sub>4</sub> plumes crossing the North Sea. In this case the data from the LRTAP sampling network are completed with concentrations measured from the NILU aircraft.

In addition, calculated concentrations are compared with observations from the LRTAP network for a period of six months starting from December 15, 1973. Based on this comparison a SO<sub>2</sub> dry deposition pattern for Europe 1974 is calculated, using computed mean concentrations and a deposition velocity of 0.8 cms<sup>-1</sup>.

### The data

The SO<sub>2</sub>-emission data used in this work are based on available information from the United Nations Economic Commission for Europe, and from OECD's Air Management Sector Group. A grid map giving the figures have been published elsewhere (Eliassen and Saltbones 1975). Better information has now been received for most of the countries, but a complete survey was not available for this investigation. The yearly emission data are believed to be within  $\pm 20\%$  from the actual figures, but may be somewhat more uncertain for the Eastern European countries. No seasonal variation has been included in the emission figures.

The air concentration measurements within the LRTAP network are carried out by laboratories in the participating countries, using sampling and analysis methods specified for the project. The detection limits have been estimated to 2-5  $\mu\text{g m}^{-3}$  for SO<sub>2</sub>, and better than 1  $\mu\text{g m}^{-3}$  for particulate SO<sub>4</sub>. Locations of the sites used in this investigation are shown in Fig 1. The geographical coordinates of the sites used in the six months comparison with model calculations, are given in Table 2.

The wind fields used for advection are based on wind observations in the 850 mb surface at 00, 06, 12 and 18 GMT. To obtain gridpoint values, the two wind components are analysed independently. The time interpolation between observation hours is linear in each component.

Description of the model

Consider a layer of air of thickness  $h$  flowing horizontally over a flat surface. Assume that the air has a constant density, that the wind does not change with height, and that the  $\text{SO}_2$  and  $\text{SO}_4$  are completely mixed up to the height  $h$ . The equations of continuity for  $\text{SO}_2$  and  $\text{SO}_4$  within the layer are

$$\frac{Dq}{dt} = E_q - F_q \quad (1)$$

$$\frac{Ds}{dt} = E_s - F_s \quad (2)$$

where  $q$  and  $s$  are the concentrations of  $\text{SO}_2$  and  $\text{SO}_4$ , and  $E_q$ ,  $E_s$ ,  $F_q$ ,  $F_s$  are source and sink terms for  $\text{SO}_2$  and  $\text{SO}_4$ . The operator  $\frac{D}{dt}$  denotes the total time derivative along a trajectory. The quantities in equations (1) and (2) are independent of the vertical coordinate.

The  $\text{SO}_2$ -emission term  $E_q$  is put equal to  $Q/h$ , where  $Q$  is the  $\text{SO}_2$ -emission per unit area and time at the current position of the trajectory, taken from the emission inventory referred to earlier. For this work, the emission map used earlier, has been transformed to another grid and extended somewhat towards the east. Both grids have a grid distance of 127 km at  $60^\circ\text{N}$ . No attempt is made to describe in detail the various transformation and removal processes of  $\text{SO}_2$ . The transformation



$\text{SO}_2 \rightarrow \text{SO}_4$  is assumed to be of first order, and the removal rates of  $\text{SO}_2$  and  $\text{SO}_4$  are assumed to be proportional to the concentrations. With these assumptions the equations (1) and (2) become

$$\frac{Dq}{dt} = \frac{Q}{h} - kq \quad (3)$$

$$\frac{Ds}{dt} = \frac{3}{2} k_t q - \kappa s \quad (4)$$

$k_t$  is the transformation rate for  $\text{SO}_2 \rightarrow \text{SO}_4$ , and  $k$ ,  $\kappa$  are removal rates for  $\text{SO}_2$  and  $\text{SO}_4$ . The factor  $3/2$  is the ratio of molecular weights of  $\text{SO}_4$  and  $\text{SO}_2$ . The following values were used for the constants:

$$k = 10^{-5} \text{ s}^{-1}$$

$$k_t = \kappa = 10^{-6} \text{ s}^{-1}$$

$$h = 10^3 \text{ m}$$

The authors have earlier (Eliassen and Saltbones, 1975) reported some estimates of  $k$  and  $k_t$  using a method based on trajectories arriving at LRTAP sampling sites. These estimates were on the average about twice as large as the values given above. When complete vertical mixing of  $\text{SO}_2$  up to the height  $h$  is assumed, a deposition velocity  $v_s = 1 \text{ cms}^{-1}$  gives a removal rate  $v_s/h = 10^{-5} \text{ s}^{-1}$ .

In the model, isobaric trajectories for marked particles are computed using the observed and analysed 850 mb winds. The  $\text{SO}_2$  and  $\text{SO}_4$  concentrations  $q$  and  $s$  associated with the marked particles change according to equations (3) and (4). At the start of the integration, the number of marked particles is equal to the number of emission squares ( $32 \times 32$ ), and each marked particle is positioned in the middle of an emission square. New positions for the particles are calculated every  $\Delta t = 1$  hr, using a method described by Petterssen (1956).

Every 12 hours, 00 and 12 GMT, the integration is restarted with new marked particles in the middle of the emission squares. By this time, about 15% of the old particles have disappeared across the grid boundary. The  $\text{SO}_2$  and  $\text{SO}_4$  concentrations of the new particles are obtained from those of the remaining old particles by an interpolation procedure, treating the directions parallel and perpendicular to the trajectories differently.

Model estimates of daily mean  $\text{SO}_2$  and  $\text{SO}_4$  concentrations at a sampling site are obtained by averaging the estimated concentrations of the timesteps covering one day. The concentration estimate at a certain timestep is the mean value of the concentrations associated with the particles present inside a circle around the sampling site with the same area as an emission square. If no particles are present inside the circle, the concentration estimate of the previous timestep is used.

An example: Computed large-scale SO<sub>2</sub> and SO<sub>4</sub> plumes verified by concentration measurements from aircraft

As part of the LRTAP programme a number of concentration measurements from aircraft have been carried out. When comparing these measurements to model estimates, one should expect the best correspondence where using measurements taken over the sea, where the anthropogenic SO<sub>2</sub>-emissions are negligible. The vertical concentration distributions may therefore closer approach the completely mixed conditions assumed in the model. This also makes the wind in the 850 mb surface a more representative advection wind for the layer. An example is shown on Figures 2 and 3.

The figures show the computed SO<sub>2</sub> and SO<sub>4</sub> concentration fields at 12 GMT May 10, 1974. A low pressure cell approaching from the west has set up a southeasterly airflow across the North Sea. The concentration measurements made with the NILU aircraft are shown on the figures, together with daily mean concentrations from the ground sampling sites of the LRTAP programme. The flight height was around 550 m.

It is seen that the model in this case gives about the right concentration levels. The observations confirm the existence of the computed SO<sub>2</sub> and SO<sub>4</sub> plumes 500 km away from the closest upwind anthropogenic sulphur emissions. Possibly a slight displacement of the computed plumes towards the left would fit

the aircraft measurements better. This is consistent with barotropic boundary layer theory since the sampling height is well below the 850 mb surface, where the winds used for advection are observed.

Model estimates compared to observed SO<sub>2</sub> and SO<sub>4</sub> air concentrations at LRTAP sampling sites

Model calculations have been carried out covering a period of more than one year, starting from December 15, 1973. The model estimates are compared with observed concentrations from the first six months of this period.

In Table 1, the computed and observed six-monthly mean values of SO<sub>2</sub> and SO<sub>4</sub> air concentrations at 29 LRTAP sampling sites are listed. The table also gives the correlation coefficients between observed and computed daily concentrations in the period. For most sampling sites the number of daily concentration pairs were between 180 and 170, except for D2, D3, DK4 where the numbers were around 160, and DK6, NL4 where they were around 150. The SO<sub>4</sub> correlation coefficients range from 0.241 to 0.775. The corresponding coefficients for SO<sub>2</sub> range from -0.019 to 0.610. At all sampling sites except two, the SO<sub>4</sub> correlation coefficients are higher than the SO<sub>2</sub> coefficients, even though the transformation SO<sub>2</sub> → SO<sub>4</sub> is described simply as a first order reaction in the model. Some explanation for this may be provided by the frequency distributions of observed and computed daily concentrations. At the site UK1

for example, (fig 4)  $\text{SO}_2$ -concentrations lower than  $16 \mu\text{g m}^{-3}$  are much more often observed than computed. The model, in which complete mixing in a grid volume is assumed, is unable to explain the observed low  $\text{SO}_2$ -concentrations in areas with large emissions. In these areas, the  $\text{SO}_2$  is far from being uniformly distributed within a grid volume, because a significant part of it is emitted from point sources, as seen from a horizontal scale of 127 km and a vertical scale of 1 km. For the  $\text{SO}_4$ , the mean transformation rate is slow enough to allow time for a more thorough mixing. Therefore,  $\text{SO}_4$  is more uniformly distributed in the atmosphere than  $\text{SO}_2$ , and behaves more according to the model assumptions.

Factors like precipitation, vertical concentration gradients and wind shear are not included in this simple advection model. This limits the day-to-day agreement obtainable between observations and model estimates.

Dry deposition of  $\text{SO}_2$  in Europe 1974 as estimated from computed mean concentrations

Figures 5 and 6 show the computed six-monthly mean concentrations plotted against the observed ones (data in Table 1). Denoting the observed and computed  $\text{SO}_2$  six-monthly mean concentrations by  $y$  and  $x$  respectively, the linear regression line of  $y$  on  $x$  is:

$$y = 0.603x + 1.85 \mu\text{g m}^{-3} \quad (5)$$

with a correlation coefficient of 0.935. Assuming random sampling from normal populations, the 99% confidence limits for the regression coefficient are  $0.603 \pm 0.125$ .

The correspondence between computed and observed  $\text{SO}_4$  six-monthly mean values is not as good (Fig 6), even though the day-to-day correlation is better than for  $\text{SO}_2$ . Evidently, the low mean values are overestimated and high ones underestimated. A larger value of  $\kappa$  in equation (4) would better this situation, as this would reduce the low computed values relatively more than the high ones. The overall  $\text{SO}_4$  concentration level can be adjusted by means of the transformation rate  $k_t$ .

The good correspondence between computed and observed six-monthly mean  $\text{SO}_2$  concentrations encourages a calculation of an  $\text{SO}_2$  dry deposition pattern in Europe for 1974. The yearly mean concentrations of  $\text{SO}_2$  for each emission square is calculated from the model concentrations at 00 and 12 GMT each day. To transform these to ground level concentrations, the computed values are adjusted by means of the line  $y = 0.719 x$ , instead of using the linear regression line (5). Both lines are shown on Fig 5. To obtain the dry deposition flux, a deposition velocity of  $0.8 \text{ cms}^{-1}$  is employed, a value estimated by Owers and Powell (1974) to be representative for the British Isles (referred to concentrations measured 20 cm over the surface). The resulting deposition map is shown on Fig 7. The map differs somewhat from the corresponding ones calculated by Bolin and

Persson (1974), with lower deposition values close to the large emissions and higher ones far away. The calculations of Bolin and Persson are based on a statistical formulation of the dispersion equation, and are valid for an arbitrary year.

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Table 1: Computed and observed six-months mean concentrations at LRTAP sampling sites. Also given are the correlation coefficients between computed and observed daily SO<sub>2</sub> and SO<sub>4</sub> concentrations in the same period, starting December 15, 1973. In addition the geographic coordinates of the sampling sites are listed. All concentrations are in  $\mu\text{gm}^{-3}$  as SO<sub>2</sub> or SO<sub>4</sub>.

	SO <sub>2</sub>			SO <sub>4</sub>			Geographic coordinates of sampling sites		
	Mean concentrations		Correlation Coefficient	Mean concentrations		Correlation Coefficient	N Lat	Longitude	Altitude (m)
	Observed	Computed		Observed	Computed				
D 2	25.8	39.8	0.194	5.2	7.2	0.241	52 48	10 45 E	73
D 3	14.6	22.9	0.033	3.9	7.5	0.354	47 58	7 57 E	1200
DK1	-	-	-	0.5	2.8	0.359	62 04	6 58 W	740
DK2	7.5	10.1	0.141	6.3	4.9	0.656	57 07	8 36 E	46
DK3	7.4	14.1	0.086	7.9	5.2	0.498	56 21	9 36 E	13
DK4	11.3	17.5	-0.006	6.8	5.2	0.447	56 00	11 17 E	3
DK5	9.2	19.2	0.321	8.0	5.8	0.484	54 44	10 44 E	8
DK6	10.3	16.9	-0.019	9.3	5.7	0.245	55 00	15 05 E	6
F 1	24.7	26.2	0.609	17.7	6.2	0.775	48 32	2 22 E	64
N 1	8.1	8.3	0.495	5.1	4.4	0.627	58 23	8 15 E	190
N 3	5.6	7.7	0.310	4.6	4.4	0.528	58 19	7 35 E	275
N 9	5.8	6.9	0.386	4.5	4.2	0.532	58 41	5 59 E	263
N22	11.5	9.4	0.229	7.0	4.2	0.489	59 04	10 26 E	35
N23	7.9	8.9	0.264	5.4	4.3	0.419	58 38	9 08 E	20
N25	3.3	3.7	0.564	1.7	3.4	0.669	62 27	11 16 E	1539
NL1	24.3	38.2	0.346	12.1	6.6	0.564	51 58	5 38 E	7
NL2	17.7	31.9	0.547	8.5	6.1	0.636	52 49	6 40 E	17
NL3	14.3	26.9	0.327	8.9	6.1	0.580	52 55	4 47 E	0
NL4*	31.5	38.6	0.568	11.0	6.6	0.661	51 28	5 29 E	29
S 3	6.2	10.0	0.444	5.3	4.1	0.501	58 46	14 18 E	125
S 4	5.1	7.8	0.030	5.5	3.8	0.495	59 46	17 05 E	30
S 5	3.1	3.2	0.194	2.7	3.0	0.435	63 51	15 17 E	405
SF1	6.5	6.9	0.284	2.2	3.7	0.479	60 11	19 59 E	15
SF2	5.2	7.9	0.242	2.4	3.6	0.363	60 49	23 30 E	104
SF3	10.5	6.6	0.486	2.9	3.5	0.390	61 34	28 04 E	120
SF4	5.8	4.4	0.498	1.9	3.3	0.427	62 31	24 13 E	154
SF5	5.1	1.8	0.318	1.4	2.6	0.340	67 22	26 39 E	178
UK1	23.4	37.0	0.236	7.7	5.3	0.700	51 58	0 06 W	125
UK2	12.7	18.4	0.610	6.0	5.0	0.689	55 19	3 12 W	236

\* The estimates are those of NL1, but fewer cases.

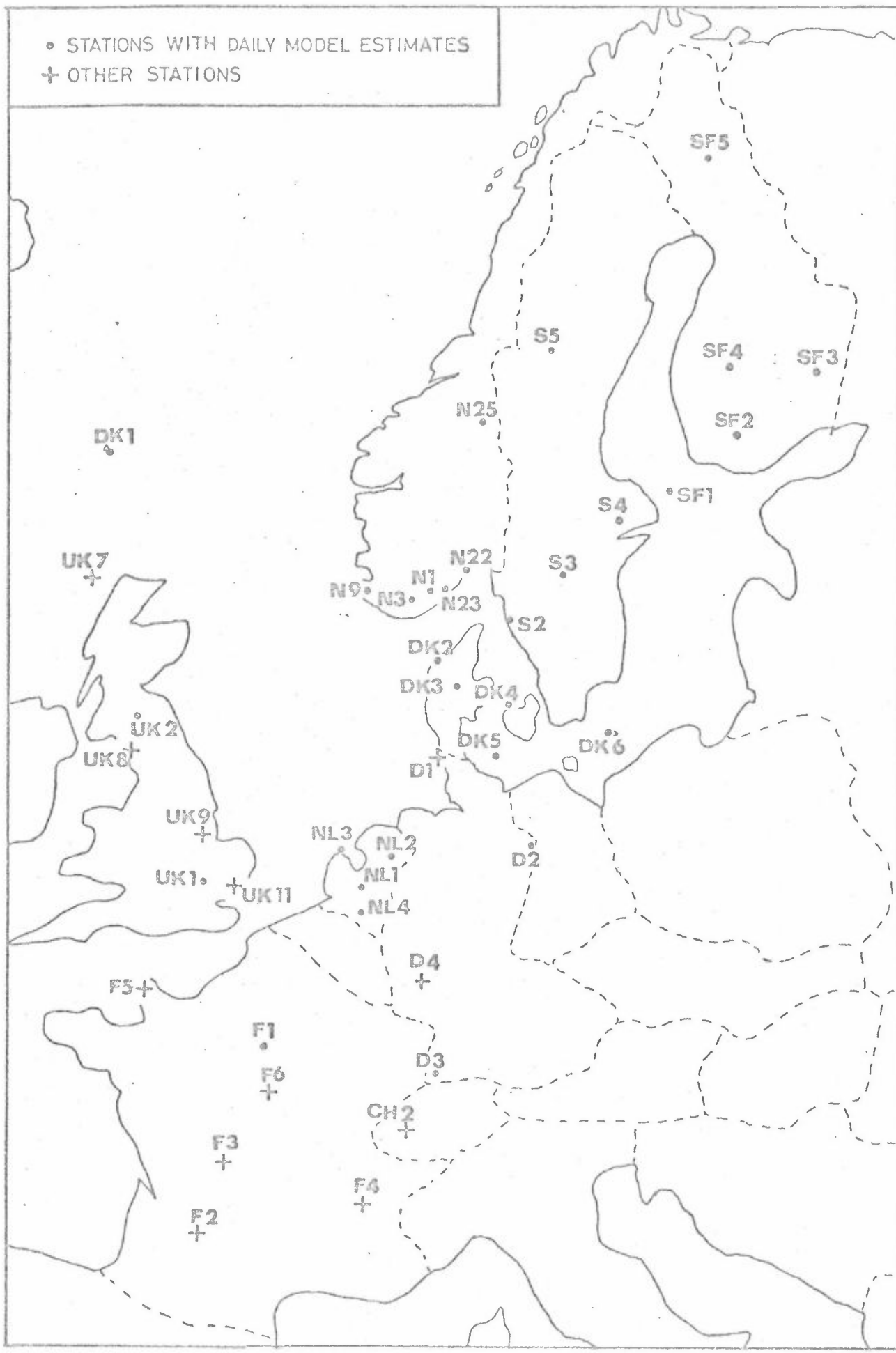


Fig 1: Location of LRTAP sampling sites providing data for this investigation. Data from stations marked with + appear only on Figs 2 and 3.

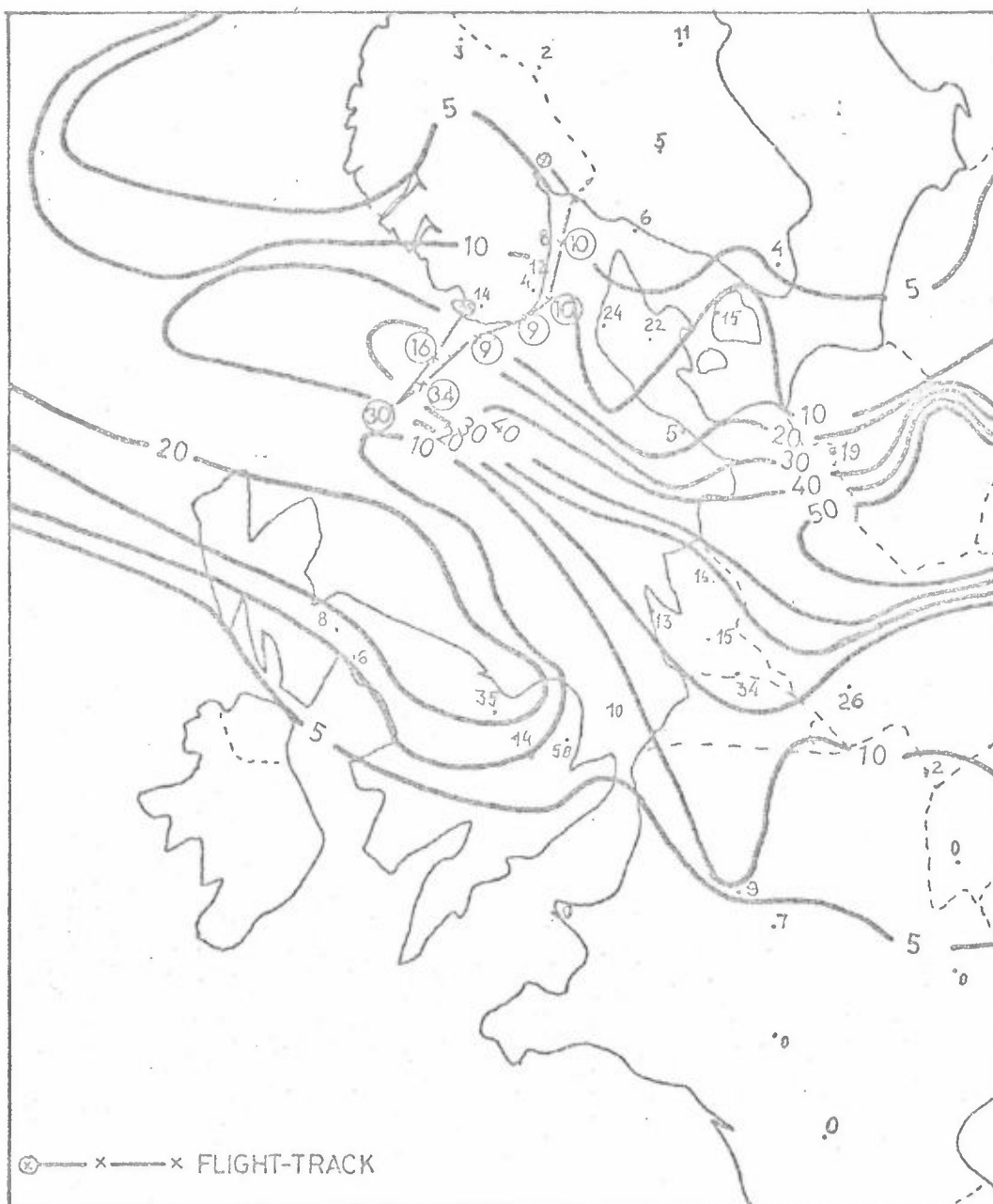


Fig 2: Computed SO<sub>2</sub>-concentrations at 12 GMT May 10, 1974, (isolines) together with aircraft measurements (in circles) and daily mean concentrations from the LRTAP-network. Unit:  $\mu\text{g SO}_2/\text{m}^3$ .

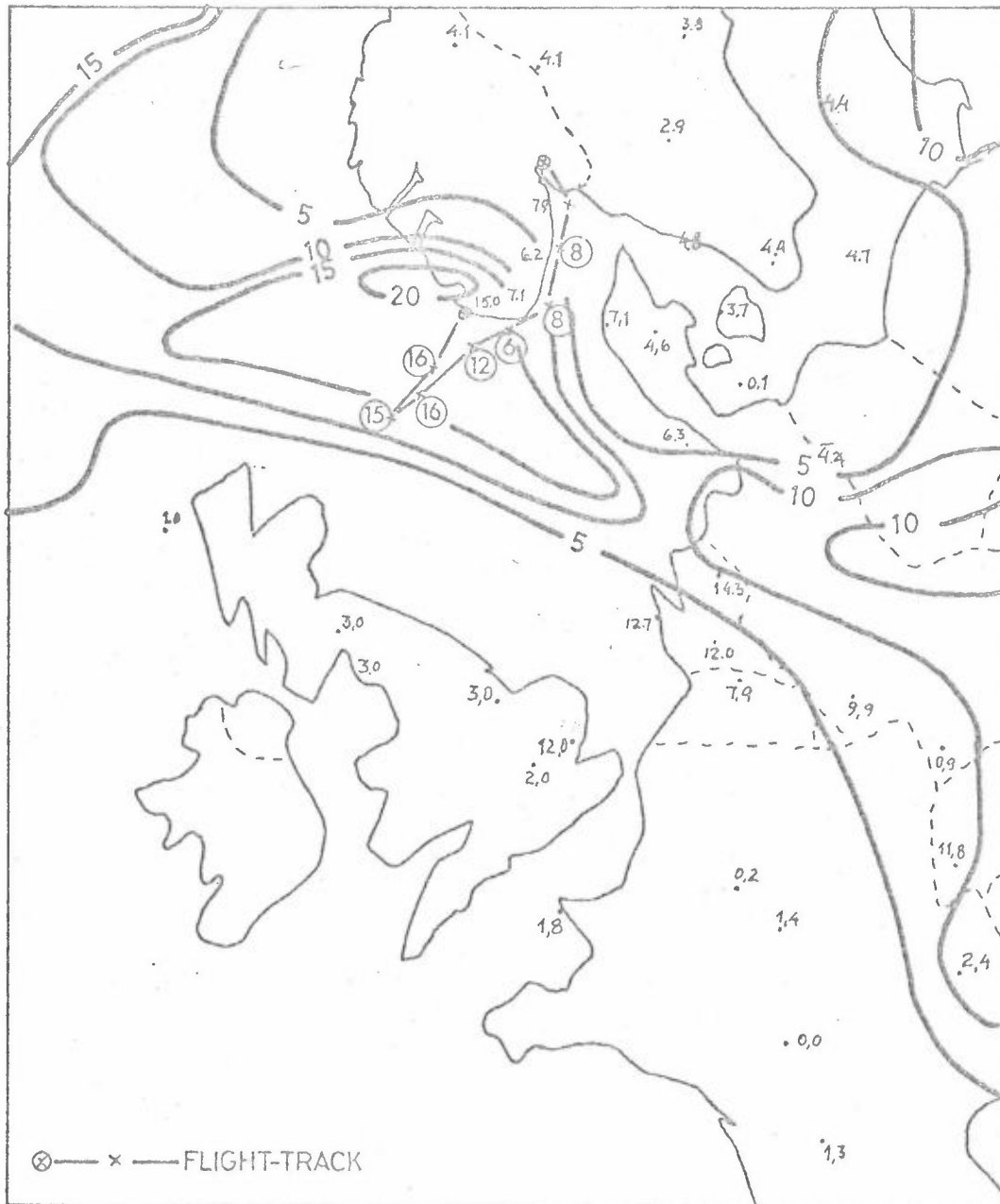


Fig 3: Computed SO<sub>4</sub>-concentrations at 12 GMT May 10, 1974, (isolines) together with aircraft measurements (in circles) and daily mean concentrations from the LRTAP-network. Unit:  $\mu\text{g SO}_4/\text{m}^3$ .

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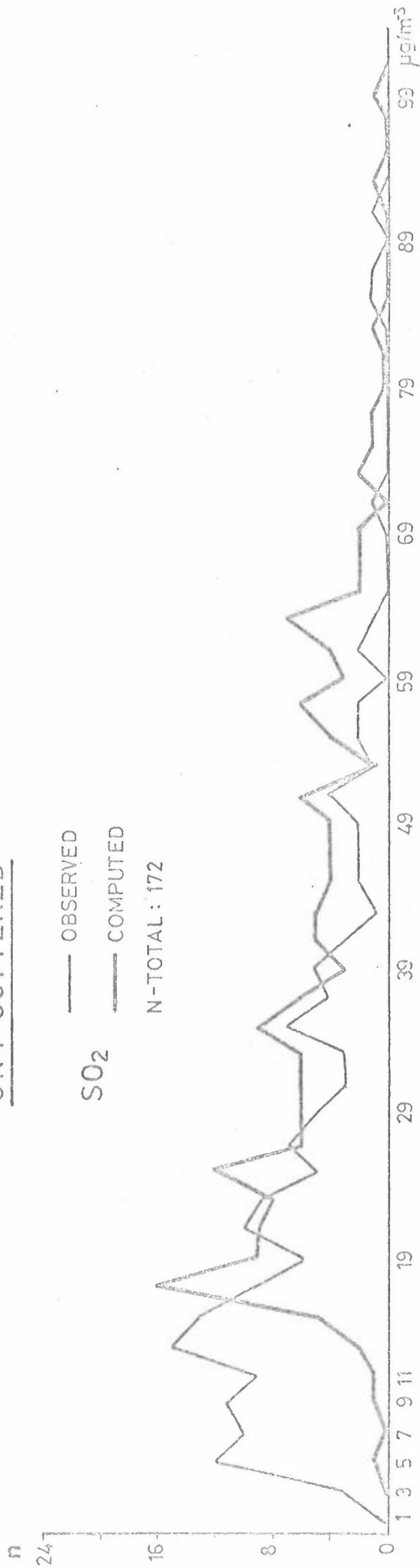


Fig 4: Frequency distribution of observed and computed daily mean SO<sub>2</sub>-concentrations at the site UK1.

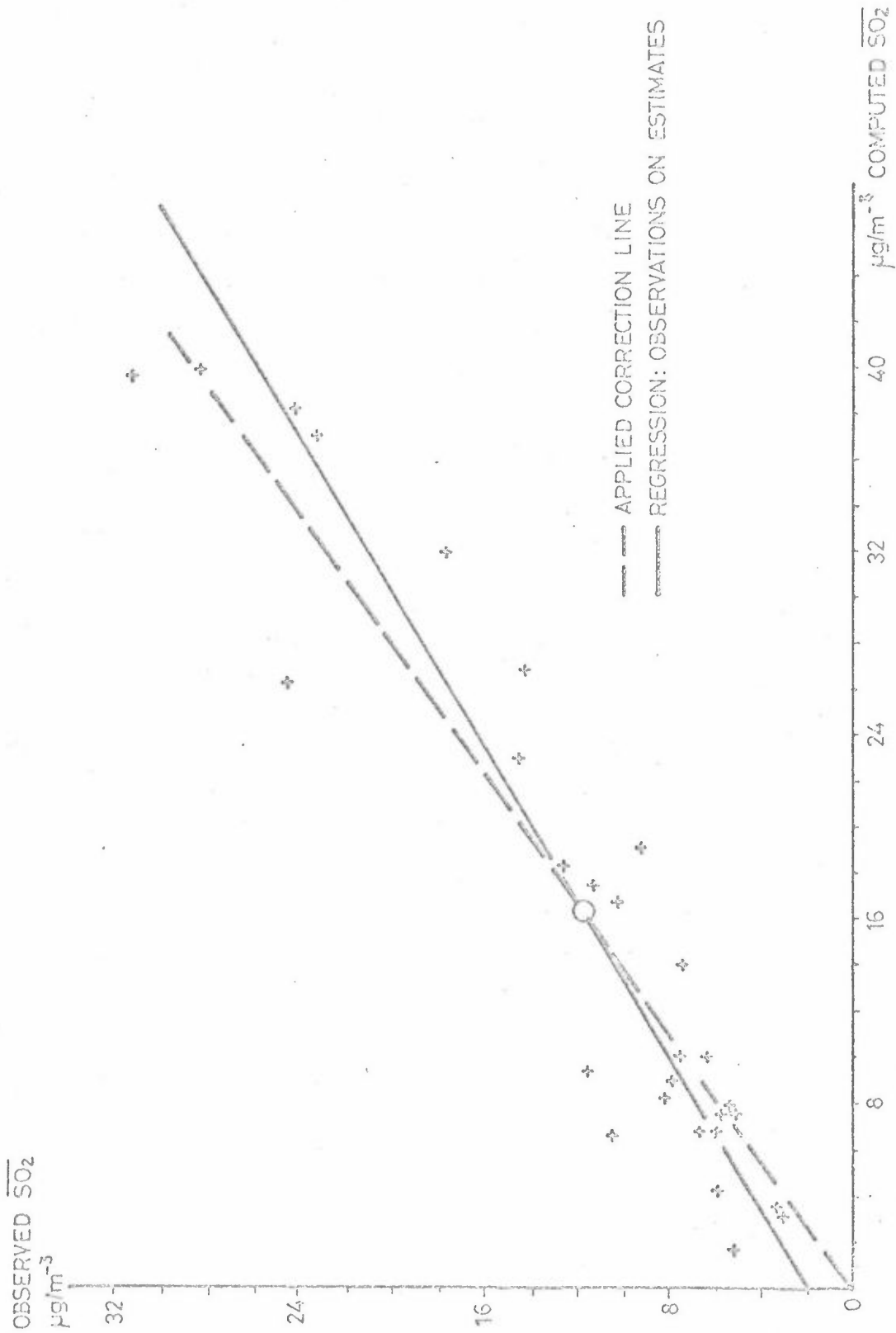


Fig 5: Computed six-monthly mean  $\text{SO}_2$ -concentrations plotted against observed ones. The linear regression line of observations on estimates is shown together with correction applied when calculating dry deposition.

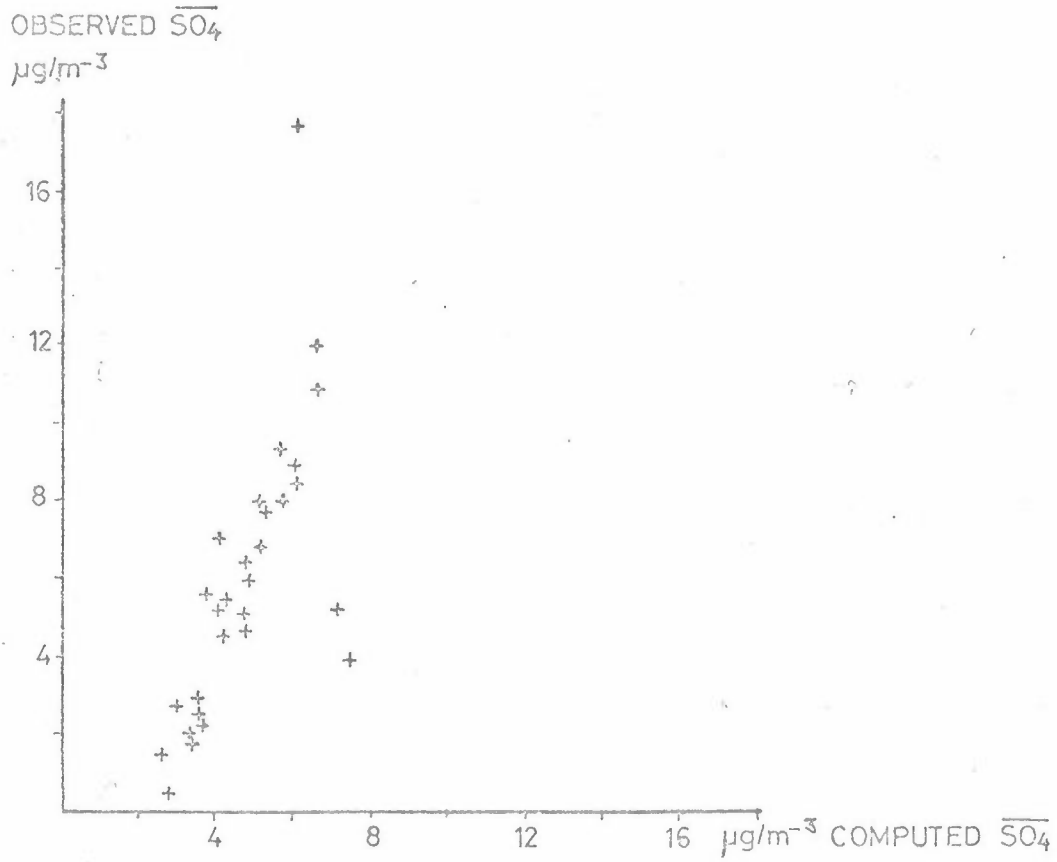


Fig 6: Computed six-monthly mean  $\text{SO}_4$ -concentrations plotted against observed ones. The linear regression line of observations on estimates is shown together with correction line applied when calculating dry deposition.





Fig 7: Calculated SO<sub>2</sub> dry deposition pattern for Europe for the year 1974. Unit: g SO<sub>2</sub>/m<sup>2</sup>.

