NORWEGIAN INSTITUTE FOR AIR RESEARCH

LRTAP 7/75

AIRCRAFT SAMPLING OF SULPHUR DIOXIDE AND SULPHATES - DISCUSSION OF RESULTS OBTAINED WITHIN THE OECD PROGRAMME

(PRELIMINARY REPORT)

Corrected edition

BY

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KJELLER 25TH APRIL, 1975

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1. INTRODUCTION

Aircraft sampling of sulphate aerosol and sulphur dioxide was part of the voluntary programme in the first measurement phase of the "Long Range Transport of Air Pollutants". The first organized samplings were performed in 1973. The second measurement phase started 1st January 1974 and terminated 31st March 1975.
The total number of known flights is 80. This report gives preliminary results from the 58 flights reported to the Central Coordinating Unit before 1st January 1975.

2. OBJECTIVES

The main objectives of the aircraft sampling programme should be (1) (2) (3)

1. To verify and correct sulphur dioxide and particulate sulphate concentration fields predicted by the atmospheric dispersion model. The measurements should represent the average concentration from the surface to the top of the mixed layer.
2. To obtain vertical profiles of pollutant concentrations under representative transport situations and allow estimates of the relative importance of sinks like dry deposition, etc...
3. To estimate the conversion rate of sulphur dioxide to sulphuric acid and sulphates by following the airstream and measure at time intervals.

Besides sampling of sulphur in particulate matter and sulphur dioxide, the particle concentration were
to be recorded using either an integrating nephelometer or a particle counter. Meteorological conditions naturally were also to be recorded.

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The Central Coordinating Unit was particularly interested in obtaining information also on nitrate, ammonium and ammonia concentration, and concentration of condensation nuclei and the chemical composition of collected droplet sampler.

Procedures for the aircraft sampling and the chemical analyses are described in (4).

Coordinated sampling flights over a large region were to be confirmed to episodes with high concentrations. These episodes were to be forecasted and the participants alerted well in advance.

Data from the measurements were to be forwarded to the Central Coordinating Unit within one month form the sampling flight.

3. PROGRESS OF WORK

As one may have expected, this rather ambitious programme could only be followed in parts. A continuous standby involving aircrafts equipped with the required instruments could simply not be kept in all the member countries. In spite of the practical difficulties, the number of flights and measurements were quite high, especially those to compare observations of sulphur with calculated concentration fields. A total of 56 episodes was forecasted until January 1975. 38 proposed flights were executed. Additional flights bring the total number to 58. Only on a few occasions did more than one country participate on the same day.

Special downwind samplings at time intervals, to estimate rates of conversion of sulphur dioxide to sulphuric acid and sulphates, were not carried out, nor was any measurement of ammonium and ammonia concentration performed.

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For details of instrumentation and sampling techniques, one will refer to individual reports from the participating countries.

In the following, all concentration values are in $\mu g/Nm^3$, abbreviated to $\mu g/m^3$.

4 RESULTS

4.1. Individual flights - an example

The NILU flight on 12 September 1974 may serve as an example of a successful flight.

Figure 1 shows the flight track and the weather conditions. Clouds covered England and Western parts of the North Sea. Along the rest of the route, it was cloudless but hazy. Sampling periods for sulphur dioxide and sulphate particles were generally 30 minutes, but 15 minutes during the vertical sampling in the North Sea.

Figure 2 shows mean values of sulphur dioxide and sulphate at sampling midpoints. The number of particles measured by a Gardner small particle detector, and the mean scattering coefficients measured with an integrated nephelometer are also given.

The scattering coefficient measurements, related to the number of particulates, indicate a general decrease of particles above the main flight level at 450 m. This is verified by the presence of thinner haze . Over Denmark, however, thick haze was observed all the way up to a diffuse upper boundary at about 1500 m.

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The non existence of a marked temperature inversion above a well defined mixing layer, were also noted on other flights, especially over sea. Unfortunately, the relative long periods required for sulphur sampling made it difficult to obtain vertical profiles and to map horizontal distribution on one and the same flight. Existence and thickness of well defined mixing layer are therefore not easily established. But visible observations seem to confirm their existence in high pressure situations with weak winds and stable air.

In Figure 3, the aircraft measurements of sulphur dioxide are compared with calculated values, and with observations (24 hourly) from the ground network. Corresponding values for sulphate are shown in Figure 4. Some deviations are large, especially for sulphur dioxide. This is a general finding from individual flights. However, differences between mean values from all flights are much smaller.

4.2. Statistics - preliminary findings

The reported 58 flights give altogether 424 measuring points, of which 408 contain sulphur dioxide and/or sulphate data. Figure 5 gives the geographical distribution of the points and Figure 6 their distribution with height. Although all data points are not statistically independent, they will be treated individually and grouped into 4 height intervals. The result is presented in Table 1. Ground values are interpolated from the station network. The calculated values are from the nearest grid point and interpolated if the difference in time exceeds 4 hours. The total number of calculated values exceeds the number of ground data. This is because the ground observations over land could not be extrapolated to give values far out at sea.

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		below 500 m	501-1000 m	1001-1500 m	ABOVE 1500 m
HEIGHT	mean	363 m	747 m	1317 m	2250 m
	S.D.	101 m	136 m	132 m	590 m
	n	132	155	105	33
SO_2	mean	21,0	14,4	8,0	9,3
	S.D.	29,6	17,0	8,1	13,2
	n	125	136	94	28
SO_4	mean	9,5	10,0	8,7	4,9
	S.D.	9,9	8,2	8,4	5,8
	n.	99	120	82	30
Total S	mean	12,6	9,5	6,3	6,0
	S.D.	15,5	8,5	5,4	7,1
	n.	97	115	75	28
SO_2 ground	mean	12,4	12,1	9,8	7,4
	S.D.	8,3	9,2	10,0	5,5
	n	110	134	86	23
SO_4 ground	mean	9,4	6,5	6,5	6,2
	S.D.	5,8	3,5	3,0	3,5
	n.	117	135	96	28
Total S ground	mean	9,1	8,2	7,0	5,3
	S.D.	5,3	5,2	5,5	3,7
	n	106	126	85	21

Table 1: Means and standard deviations (S.D.) within selected height intervals. Concentrations in $\mu g/m^3$. n: number of data points -

In Table 2, mean values and standard deviations of all observations, regardless of height, are shown:

	AIR			GROUND			CALCULATED		
	SO_2	SO_4	Tot. S	SO_2	SO_4	Tot. S	SO_2	SO_4	Tot. S
Mean	14,6	9,1	9,4	11,4	7,4	8,0	19,6	6,7	12,2
S.D.	21,1	8,7	10,9	9,0	4,5	5,3	16,8	8,8	9,6
n	383	331	315	353	377	338	424	424	424

Table 2: Means and standard deviation (S.D.) of total number of observations. Concentrations in $\mu g/m^3$. n: number of data points.

4.3. Mean vertical profiles

Using the mean values in Table 1 and Table 2, vertical profiles can be drawn. Figure 7 shows the distribution of sulphur dioxide. At ground level, the value given in Table 1 corresponding to aircraft observations below 500 meters is used. This is believed to give the most realistic gradient. It is also done for the sulphate profile in Figure 8, and for the total sulphur profile in Figure 9.

The sulphur dioxide curve shows a marked maximum at a height of a few hundred meters. This, however, does not necessarily apply to every individual profile. In fact, only about 50 % of the observed values at or below 500 meters were higher than the corresponding ground values. It seemed to occur mainly for the higher concentrations. High concentrations are linked with weak winds and light turbulence which again favour large gradients. Some gradients may therefore have been considerably greater than indicated in the figures.

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Any clear correlation with stability, expressed by the Richardson number, could not be detected. Temperature and wind differences between the 850 mb level and the 10 m level were used to calculate this parameter.

A calculated concentration C_{cal} represents a mean value:

$$C_{cal} = \frac{1}{H} \cdot 10^{-3} \int_0^\infty C dz$$

where C is the concentration and H = 1000 m. The figures show that the calculated values are too high, although for sulphur dioxide it agrees remarkably well with the mean value at 500 m. The figures also indicate a too slow transfer of SO_2 to SO_4 in the model.

With respect to sink mechanisms, dry depositions to the ground may be considered. Assuming a constant vertical flux in the lowest few hundred meters, and neglecting advection, we have:

$$C_0 \cdot v_d = K \cdot \frac{dC}{dz} ,$$

where v is the deposition velocity, K the turbulent eddy diffusitivity, and C_o the ground level concentration.

with $\frac{dC}{dz} = 8/200\ \mu g/m^4$, $C_o = 12\ \mu g/m^3$ and $K = 3\ m^2/s$

as a reasonable value, v_d becomes 0,010 m/s . This mean value is frequently quoted. In individual cases, the deposition velocity will vary according to stability conditions.

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CONCLUSION

1. There seems to be good agreement between observed mean concentrations at the 500 m level and calculated values. This will probably be the case also when the rest of the aircraft data are included. They contain more than 110 additional data points. However, the individual flights show considerable variation between observed, calculated and ground level observations.

2. The episodes occured mainly in weather situations with relatively weak winds, stable air and high concentrations of sulphur compounds. Maximum concentrations were observed at height levels of the order of a few hundred meters. This may not have be the case if all types of weather had been included.

3. The existence of a well mixed layer with constant concentrations below a well defined upper lid is not a general rule. Concentrations seems to decrease gradually with height above the maximum level.

REFERENCES

(1) Aircraft measurements, present status and plans, LRTAP 1/73, September 1973

(2) Proposed plan for the second measurement phase, aircraft measurements, Addendum III to NR/ENV/73.53, OECD.

(3) Meeting on the correlation of aircraft sampling, Bilthoven, 15-16 January, 1974, LRTAP 30th January 1974.

(4) Procedures for aircraft sampling and chemical analysis, LRTAP 2/73, 3 December, 1973

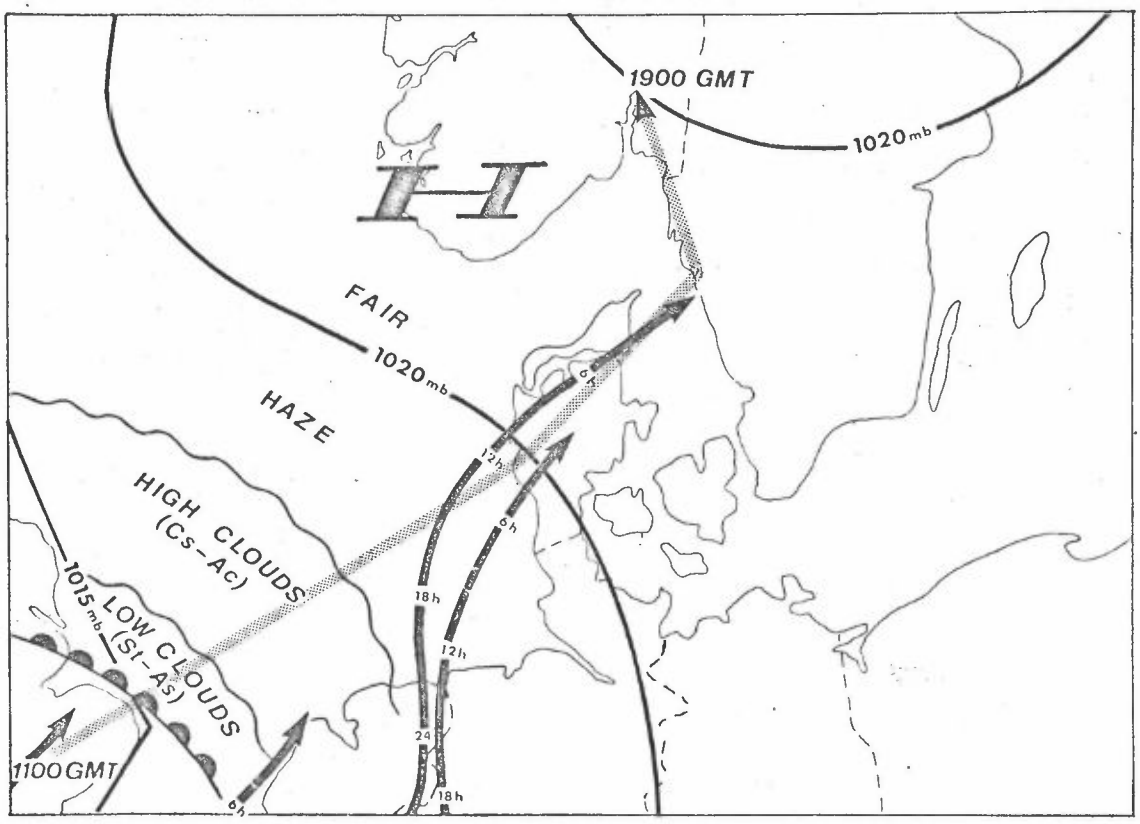

Figure 1: Flight track and weather on 12th September, 1974. Isobars and air trajectories at the 850 mb level, 1200 GMT

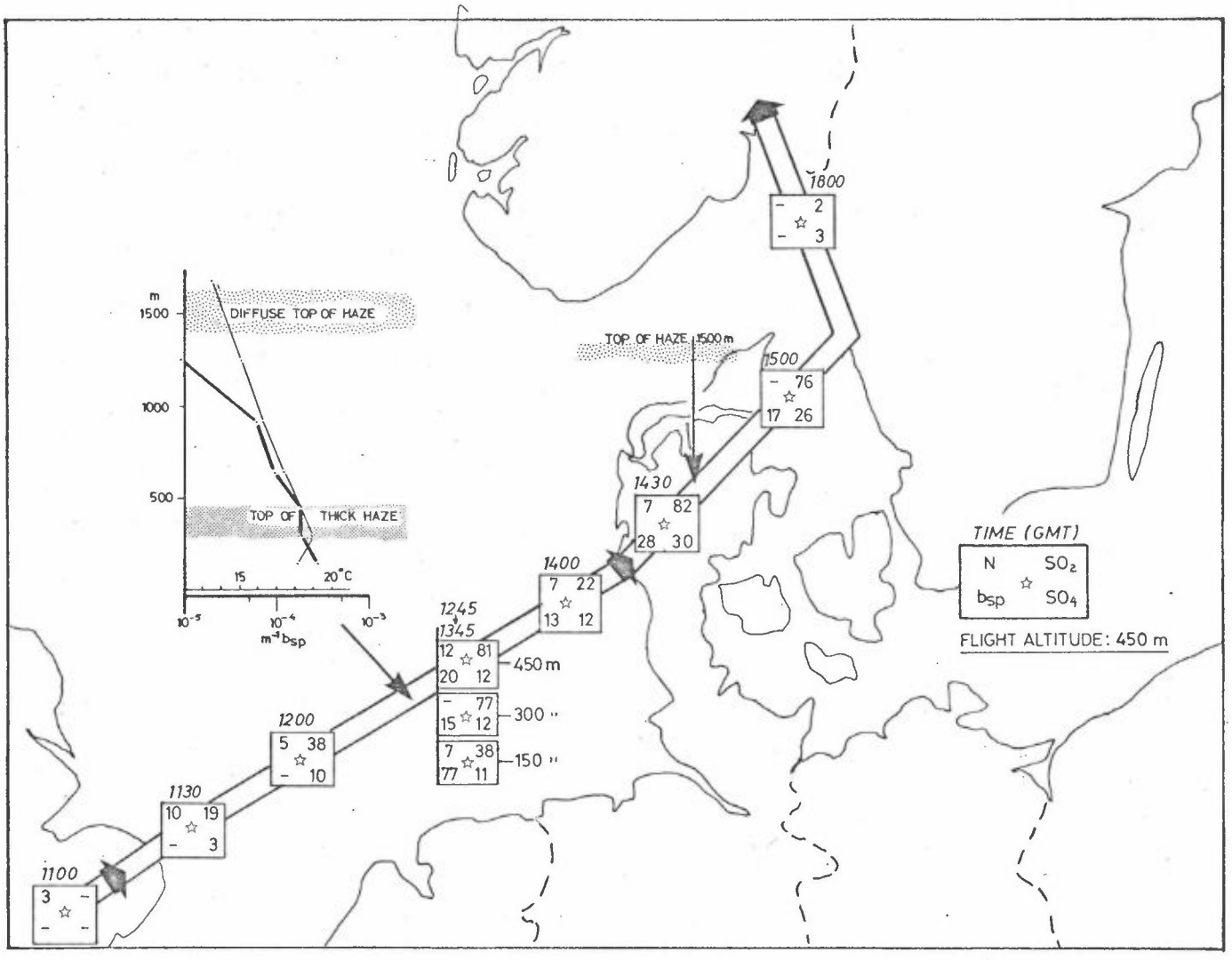

Figure 2 : Observed concentrations of SO_2 and SO_4 in $\mu g/m^3$ on 12th September, 1974.
Vertical profiles of b_{sp} and temperature.
N: number of small particle per $cm^3 \times 10^3$.
b_{sp}: scattering coefficient in $m^{-1} \times 10^{-5}$

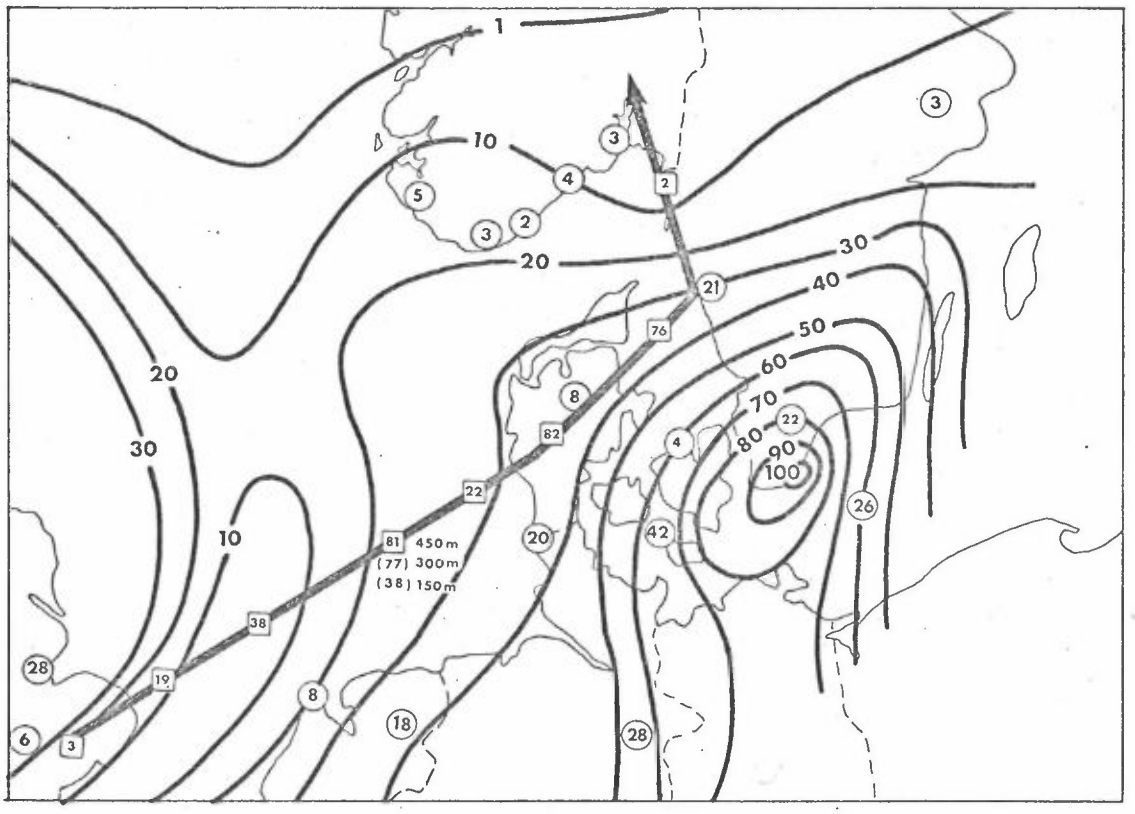

Figure 3: Sulphur dioxide concentrations in µgm/m^3, 12th September, 1974.
Isolines: Calculated concentrations,
In squares: Aircraft observations
In circles: Ground observations (24 hours).

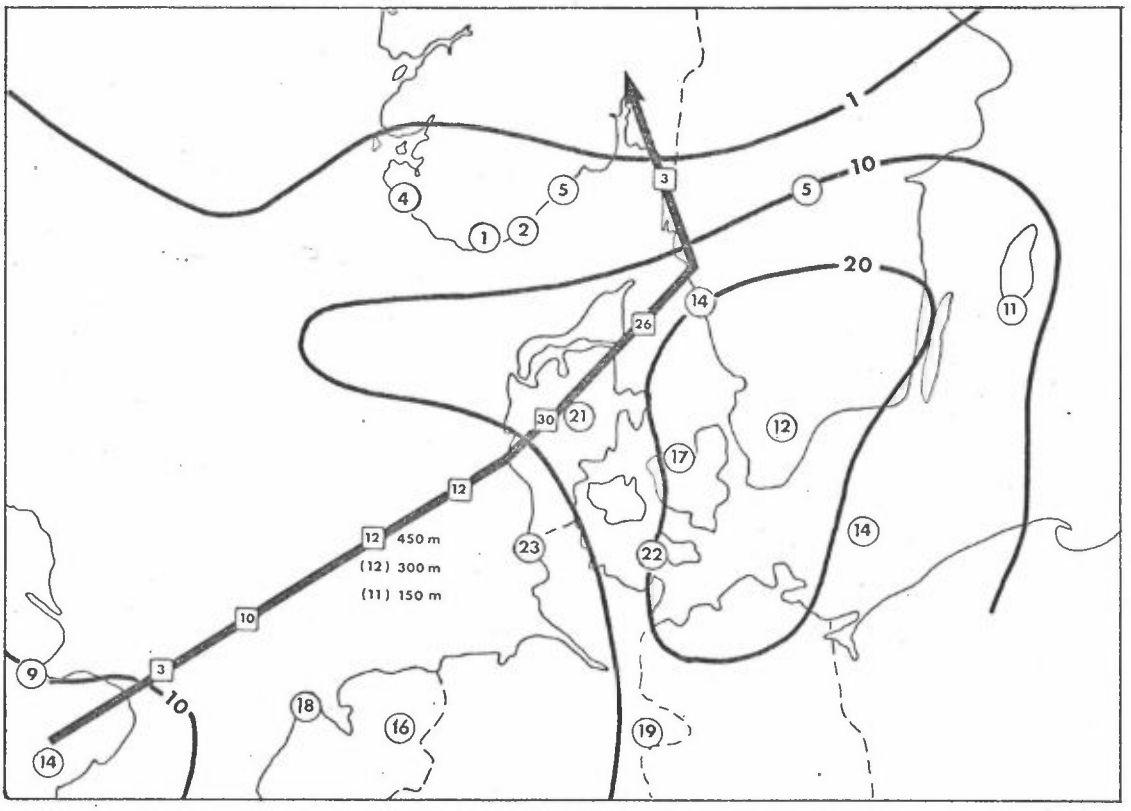

Figure 4: Sulphate concentrations in µg/m^3, 12th Sept., 1974

Isolines: Calculated concentrations

In squares: Aircraft observations

In circles: Ground observations (24 hours)

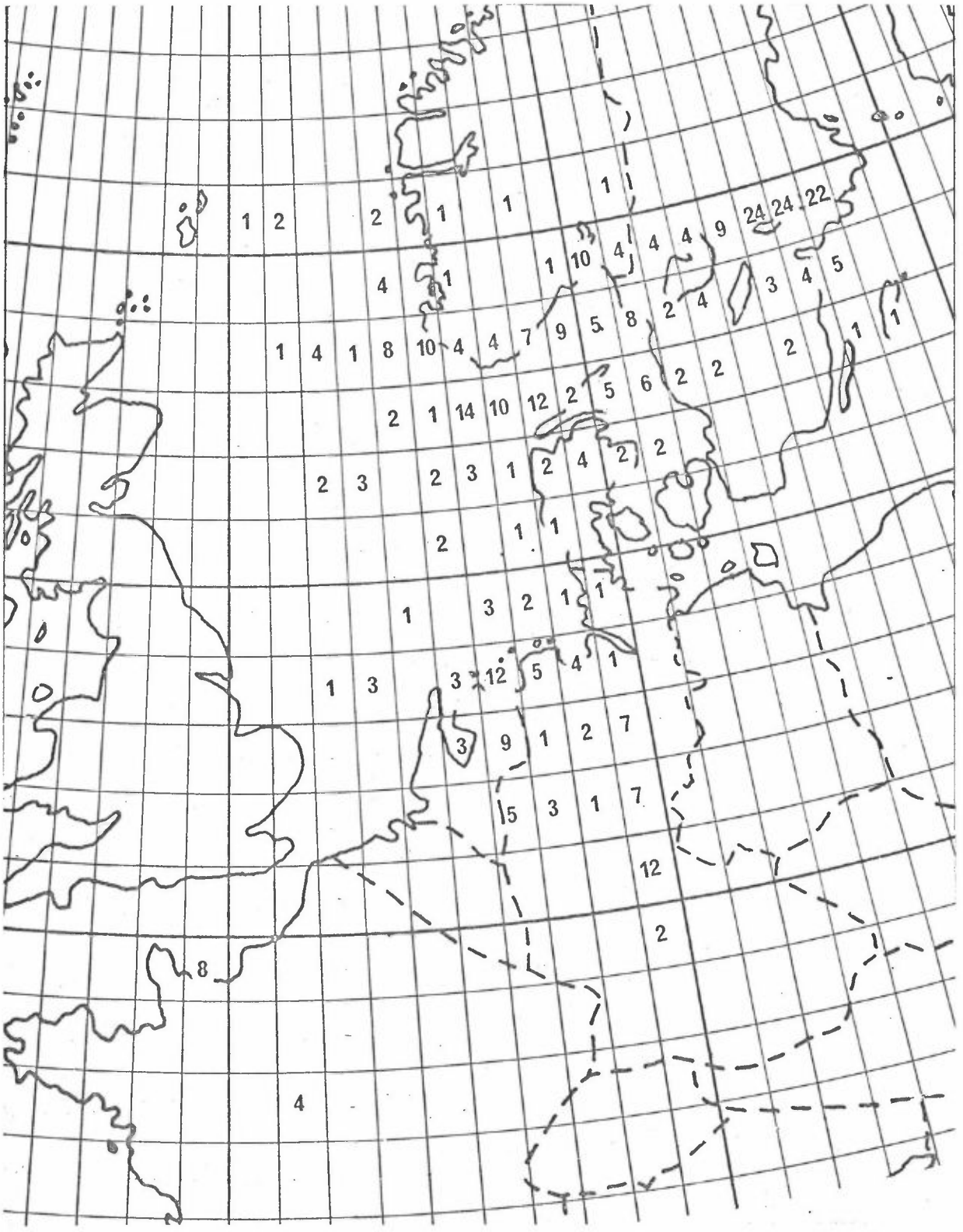

Figure 5: Geographical distribution of data points.

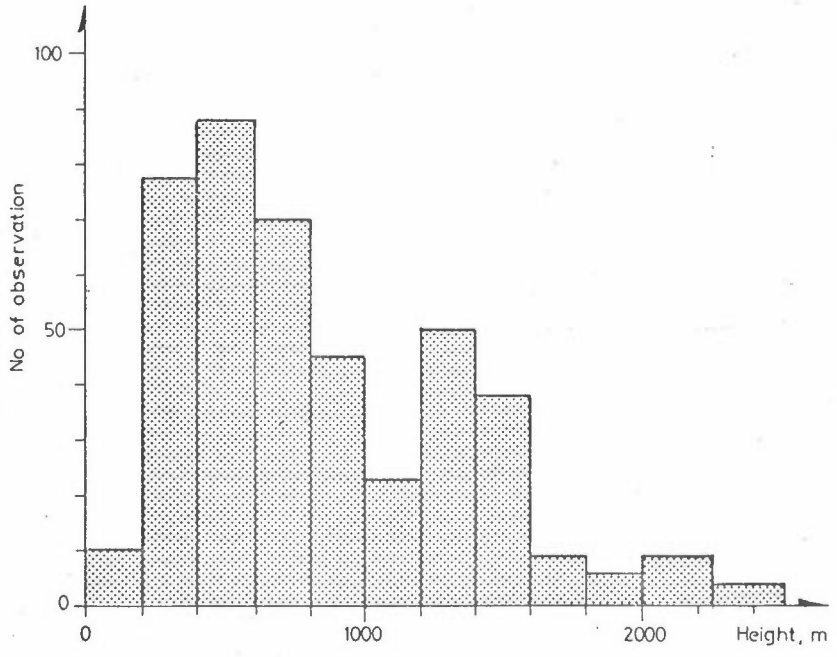

Figure 6: Number of data points within 200 m height intervals.

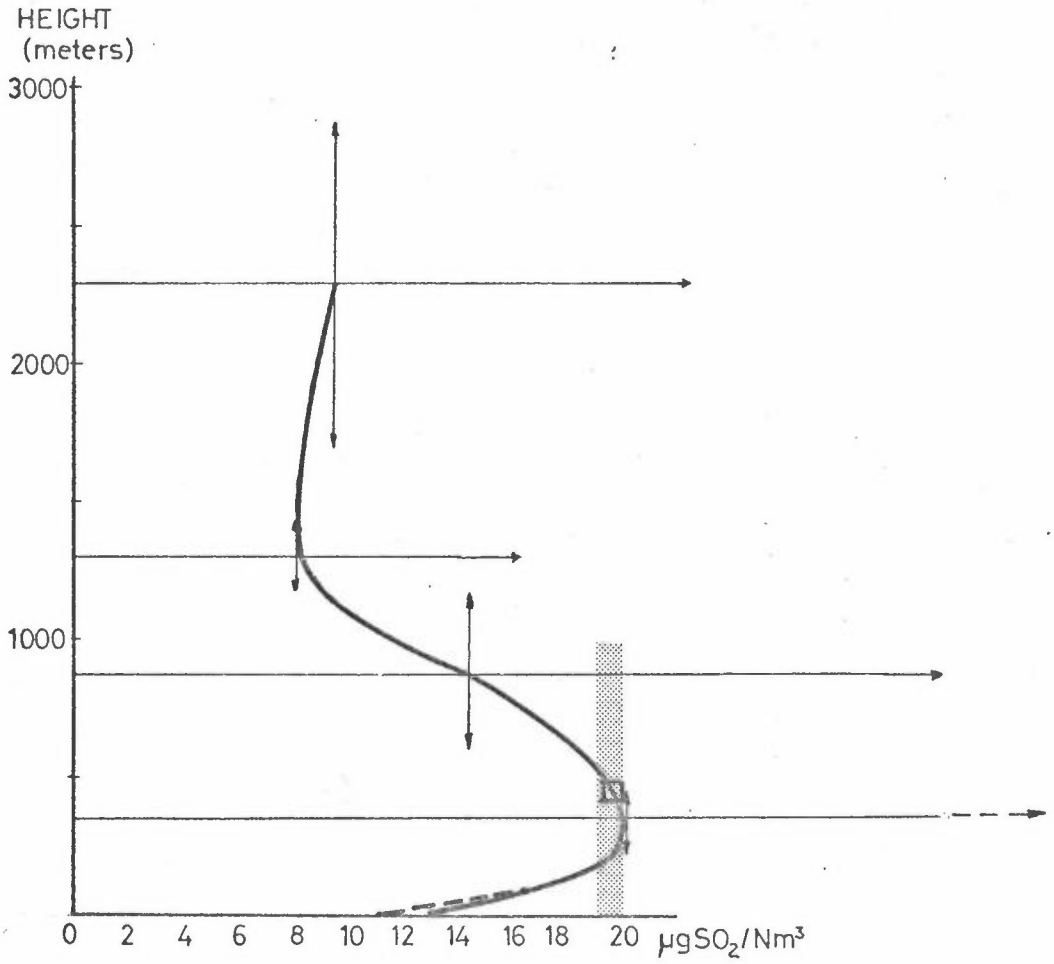

Figure 7: Mean vertical distribution of sulphur dioxide. Standard deviations of heights and concentrations shown by arrows.
Full line: Ground value from table 1, corresponding to data points below 500 m. Mean calculated value in square.
Dotted line: Ground value from Table 2, all observations

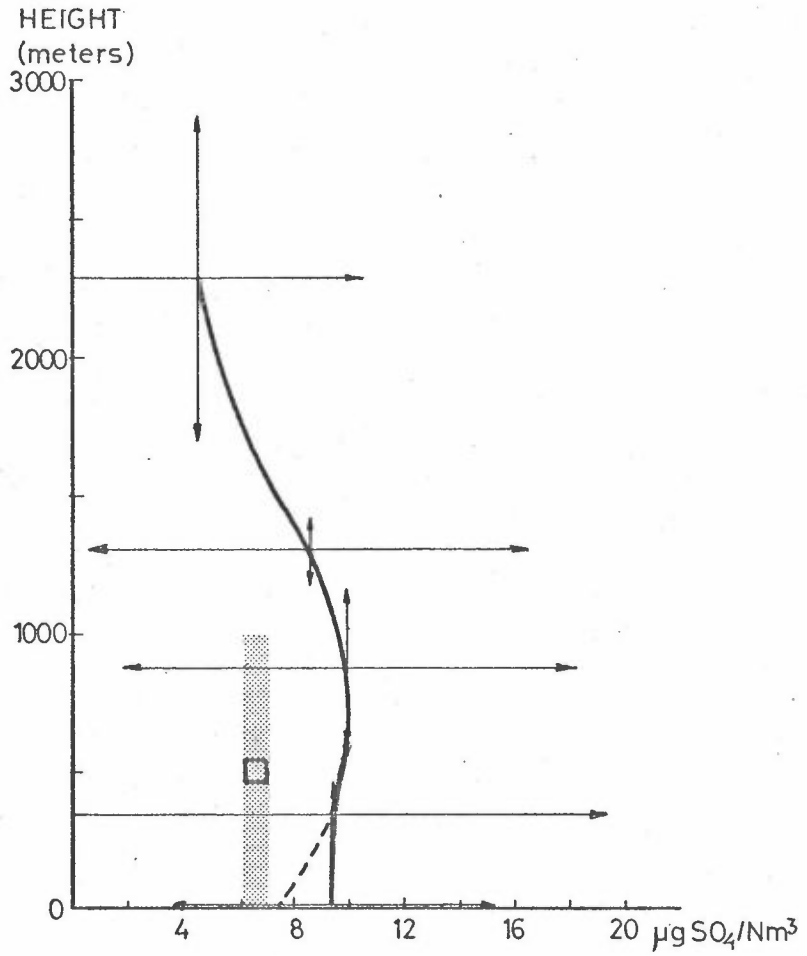

Figure 8: Mean vertical distribution of sulphate.
Standard deviations of heights and concentrations shown by arrows.
Full line: Ground value from table 1, corresponding to data points below 500 m. Mean calculated value in square.
Dotted line: Ground value from table 2, all observations

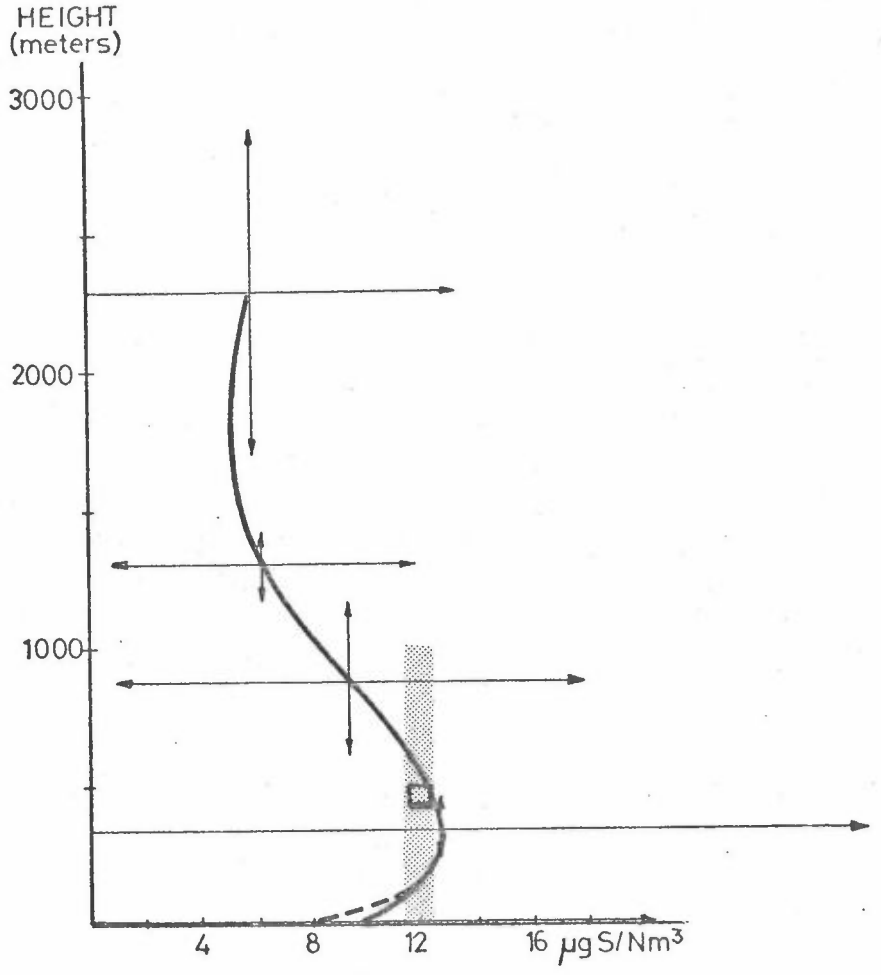

Figure 9: Mean vertical distribution of total sulphur concentrations.
Standard deviations of heights and concentrations shown by arrows.
Full line: Ground value from table 1, corresponding to data points below 500 m. Mean calculated value in square.
Dotted line: Ground value from table 2, all observations.