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METEOROLOGICAL ASPECTS OF THE TRANSPORT OF POLLUTION OVER LONG DISTANCES

by

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Meteorological aspects of the transport of pollution over long distances. By SMITH,F.B. and HUNT,R.D.

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LIST OF SYMBOLS

c	concentration (μg m^{-3})
$\bar{c}_2$	average concentration of SO_2 on sampling track
C	convergence
D_2	amount of SO_2 lost by deposition
E_2	estimated emission flux of SO_2 (t h^{-1})
f_p	frequency of stability categories
F_i	percentage lost by dry deposition minus that given by box model (%)
i	number of gridlengths downwind from source
l	length of side of grid square
M_2	measured flux of SO_2 on sampling track
P	Pasquill stability category
Q	area source strength
r	radius of active cell
R	rate of rainfall (mm h^{-1})
R_4	amount of sulphate removed by rain
S_4	production of sulphate from SO_2
S_4^*	production of sulphate implied by empirical curve (Figure 3)
t	time of travel from source
v_d	velocity of deposition (cm s^{-1})
$\bar{V}$	mean speed of sampled air (m s^{-1})
V_G	geostrophic wind speed (m s^{-1})
V_p	mean plume wind speed (m s^{-1})
V_0	surface (10 m) (m s^{-1})
V_{900}	900 mb wind speed (m s^{-1})
W_i	weighted average loss (%)
x	distance from source (km)
z_i	height of boundary layer (m)
z_{im}	median height of inversion (m)
$\bar{z}_i$	mean height of inversion (m)
$\bar{\theta}$	mean direction of wind between surface and 900 mb
θ_G	geostrophic wind direction
θ_p	mean plume wind direction
θ_0	surface wind direction
θ_{900}	900 mb wind direction
λ_i	a coefficient
Λ_i	the sum of the enhanced ground level concentration minus the concentration given by the box model
σ_y	horizontal spread of pollutant (km)
σ_z	vertical extent (r.m.s.) of plume along track (m)
GMT	Greenwich mean time
MST	mean solar time

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ABSTRACT

Since sulphur pollution is carried by the atmosphere over considerable distances it is important to have an adequate understanding of those aspects of the atmosphere's structure which are important in this process and to know not only the mean conditions but also have a good feel for the statistical variations and their significance to both wet and dry depositions to the ground. This paper discusses several aspects of the role of the atmosphere and starts by considering the nature of the mixing layer and gives statistics of its depth from measurements made at Cardington, U.K.

It goes on to summarize conclusions reached from aircraft sampling-flights over the North Sea (1971-75) in terms of the velocity of deposition of SO_2 (0.8 cm s^{-1} over land, 0.5 cm s^{-1} over sea) and the conversion rate of SO_2 to sulphate (roughly 1% h^{-1}). The next section considers particular states of the atmosphere which can lead to episodes of very high deposition of sulphate in precipitation. Such episodes are given a formal definition and their cause and geographical distribution in western Europe are investigated. Two case-studies of notable episodes are presented and a tentative conclusion reached that average statistics of the state of the atmosphere should be used only with caution when modelling long-term pollution transport in view of the relative dominance of episodes and their own rather distinctive meteorology.

Finally the paper reviews some of the rather more important aspects of meteorology which have a direct bearing on transport and which require further theoretical and experimental study.

1. THE NATURE OF THE BOUNDARY LAYER AND ITS EVOLUTION

The transport of any pollutant such as sulphur dioxide over long distances in the atmosphere is strongly influenced by the time-dependent depth of the layer over which it is dispersed. The deeper the layer, the lower the conversion rate to sulphate (other things being equal) and the more likely wind-direction variations with height will be important in spreading out the plume in the horizontal lateral direction.

This layer is often called the mixing layer and depends on the existence of turbulent eddies capable of transporting the pollutant through the vertical. This dependence on the vertical structure of turbulence equates the mixing layer to the meteorologically defined boundary layer. The latter is the lowest layer of the atmosphere under direct influence of the underlying surface (Smith and Carson, 1977). The sources and sinks of turbulent energy which characterize this layer of variable depth all result directly or indirectly from the interactions between the atmosphere and the surface. These sources and sinks are :

(a) The removal of momentum by viscous drag, the so-called shearing stress, which provides a source of turbulent kinetic energy not only very near the ground but at all levels wherever the momentum loss is effective in creating velocity shear.

(b) The input to, or extraction from, the atmosphere of sensible heat resulting from a net imbalance of radiational fluxes to and from the surface giving a temperature difference between air and surface. The flow of heat results in density differences; when heat flows into the air, buoyancy forces provide a source of turbulent energy and the boundary layer is 'unstable' whereas when heat is extracted, as on a calm night, the direction of the buoyancy forces is reversed and any turbulent motions are damped and the layer is then 'stable'. A stable boundary layer is clearly not a mixing layer in the limit when turbulence is completely quenched.

(c) The input of moisture by evaporation from the surface also creates buoyancy forces which are a source of turbulent energy. Condensation may occur at some level and when it does so not only may radiational cooling effects be important (Deardorff, 1976) in the local energy balance but latent heat of vaporization will be released.

(d) Radiation may cause warming and cooling at any level and although this effect is not restricted to the boundary layer it has to be allowed for on occasions in the overall balance.

The boundary layer then can be either unstable if the sources of energy outweigh the sinks or stable when the reverse is true. In the unstable situation the boundary layer extends up to a height characterist ically between 400 metres and 2000 metres (see later for more details). The turbulent energy does not continue to increase indefinitely of course but is cascaded from the larger eddies to the smallest eddies where viscous dissipation returns the energy to heat. This separation of wavelengths between input and output of energy is equally true in the stable situation and means that even in the latter case when the sink of energy is very strong some vestige of turbulence (and hence mixing) can exist. The stable layer, slightly turbulent or quiescent, may be only a few tens of metres deep to about 400 metres deep.

The most important question we face is how to define the upper limit of the boundary layer. This is a surprisingly difficult question to answer. Sometimes it is obvious; a very well-marked inversion at several hundred metres above ground capping a well-mixed layer provides an indisputable upper limit to the boundary layer, everything above is unaffected in a direct sense by the underlying surface whereas everything below is under its influence. On other occasions, however, no clearly marked top exists and the effect of the surface decreases only very gradually with increasing height. This is often true when buoyancy forces are very nearly zero when the boundary layer is called 'neutral' a transitional state between an unstable and a stable regime.

Perhaps the best we can do is to define the turbulent boundary layer as that part of the atmosphere extending from the underlying surface up to a height at which all turbulent and radiative flux-divergences resulting from surface action have fallen to some prespecified small percentage (say 1%) of their surface values. However, sometimes even this simple notion is confused by processes such as penetrative convection where a lower layer, in which the influence of the underlying surface is felt in a widespread direct and continuous way, is overlaid by an upper layer influenced locally and spasmodically in, say, deep cumulus clouds but otherwise uninfluenced and non-turbulent. Rather arbitrarily we restrict the boundary layer to cover the lower layer only.

To sum up then, the true boundary layer is a region dominated by a balance of turbulent-energy sources and sinks all derived from the interactions of atmosphere and underlying surface. It has a variable depth ranging typically from a few tens of metres in stable conditions to about 2000 metres in very unstable situations. The layer is characterized by significant vertical flux divergences of momentum and frequently of heat and moisture. The wind speed tends to fall below its free-stream value found above the layer, eventually to zero at the surface although the fall-off may not always be monotonic, especially in stable conditions. Wind direction also changes with height with the biggest deviation from the geostrophic direction occurring at the surface, and whilst the deviation tends to be quite variable, influenced as it is by changes with time in stability, by local topography and meso- and synoptic-scale accelerations, on the average the surface wind is backed some 5° over the sea and 10° over land in unstable conditions and 30° or more in stable conditions. The resulting flow down the pressure gradient provides a significant source of momentum to balance the shearing stress of the ground and on a synoptic scale is important in generating horizontal convergence and upward motions. The turning is also potentially important in enhancing lateral dispersion of plumes coming from major source regions of sulphur pollution.

The boundary layer frequently plays another role, that of a buffer zone into which the heat, moisture and pollution coming from sources at the ground are stored until they are released at a later date into the main body of the atmosphere. Typically in temperate latitudes the boundary layer takes several days (2-4 days) to reach this 'break-down' situation. Break-down may occur as a result of large-scale convective instability, at fronts when upsliding occurs, in mountainous regions where rotors and 'hydraulic jumps' may cause considerable vertical stirring, and by continuous synoptic-scale vertical motion aided by the diurnal cycle in the boundary layer which 'pumps' the contents of the boundary layer upwards in the middle of the day, leaving it behind thereafter to be acted upon by the steady synoptic upward motions.

Boundary layers are almost always in a state of change; they are either responding to a change in the synoptic pressure field or to some change in the underlying surface. For gradual changes the structure of the layer maintains a quasi-equilibrium but coast lines, sharp mountains and other sudden discontinuities pose very special problems of adjustment. Fortunately these changes do not normally have a very pronounced effect on the main factor affecting the long range transport of pollution; namely the mixing depth, or boundary layer height z_i .

The mixing depth may be estimated in the following ways :

(a) When z_i is relatively small, as at night, turbulence probes, or sensitive anemometers, situated on tall masts can indicate at what level the vertical mixing process ceases.

(b) Temperature and humidity profiles obtained from instruments supported on the cable of a tethered-balloon which is systematically raised and lowered can clearly indicate the change-over from boundary layer conditions to 'free' atmosphere conditions. The so-called BALTHUM operated at Cardington in the United Kingdom is a good example of this system.

(c) Radiosondes perform a very similar function although with slightly less accuracy, and are invaluable for monitoring higher inversions capping the boundary layer, beyond the normal reach of tethered balloon systems.

(d) In the absence of synoptically induced capping inversions the evolutionary induced inversions produced at the top of the layer are weaker and rather harder to detect in only moderately unstable conditions. Boundary layer theory can then be useful to complement any measurements that are available and help to decide which of perhaps several slight changes in lapse rate really marks the true top of mixing. Allowing for all the assumptions and approximations inherent in any scheme that is based on very limited input data, Figure 1 shows a simple nomogram for estimating z_i in day-time conditions based on time of day, month, cloud cover and wind speed at 10 metres. Essentially it says that the first three parameters determine the integrated heat input into the boundary layer from dawn. This heat will have modified the vertical temperature profile most likely to have been associated with that cloud cover (assumed constant in time), and time of year, based on a very detailed analysis of BALTHUM ascents made by Hardy (1973), (internal Met Office Note). The nature of the modification is according to the theory of Carson (1973). The nomogram also recognizes the two main sources of energy for boundary layer development: the generation of turbulence by the wind and the influence of sensible heating.

(e) In statistical modelling of the long-range transport of pollution it may be sufficient to know the statistical behaviour of z_i as a function of time of day. Hardy (1973) has analysed over 4000 ascents made by the BALTHUM system. Table 1 gives the essential statistics for near neutral or unstable boundary layers.

TABLE 1 Percentage of ascents showing an inversion (intensity $\geq 0.5°C$) within the indicated height range.
z_{im} is the median inversion height i.e. on 50% of all occasions an inversion lies at or below this height.)

Time GMT	z_i (m) 0-1	1-75	75-300	300-600	600-900	900-1200	Percentage with inversions at top of mixing layer	Height of inversion mean $\bar{z}_i$	median z_{im}
	%	%	%	%	%	%	%	m	m
00	59.5	9.5	5.8	3.5	2.8	2.2	85	100	0
06	32.9	12.2	16.2	6.1	4.3	2.9	78	200	130
12	2.4	5.6	4.5	8.1	9.7	5.2	50	800	1700
18	34.2	4.6	3.0	2.8	3.5	3.7	58	200	1000
All hrs	33.2	8.0	7.6	5.0	4.4	4.0	–	–	–

Considering the mean inversion height when a capped layer exists, at midday $\bar{z}_i$ = 800 metres. However, a strong seasonal trend is very clear; during the three summer months $\bar{z}_i$ = 1300 m, whereas during winter $\bar{z}_i$ = 500 m.

Equally important are stable boundary layers where the inversion is at ground level. Although incorporated in Table 1, Hardy makes a special study of them and his results are summarized in Table 2. They are particularly relevant to the long-range transport problem because they 'insulate' the main body of the pollution cloud from the effect of ground absorption (dry deposition).

TABLE 2 Details of surface inversions at Cardington obtained from BALTHUM ascents (after Hardy, 1973).

Time GMT	Number of ascents	Number of occasions of no inversions at ground	Number of occasions of inversions at ground level with top below 900 m	above 900 m	Mean height of inversion top
		%	%	%	m
00	1972	30.5	67	2.5	170
06	2074	55.4	43	1.6	160
12	1695	89.5	10.4	0.1	30
18	1847	51.4	48	0.7	70

From what has already been said it is clear that the evolution in time of the boundary layer and its variation from unstable well-mixed conditions to stable almost quiescent conditions and back again is a very important phenomenon. Figure 2 is an example of such a diurnal cycle and shows the response of the boundary layer to variations in surface sensible heat flux. Before sunrise the heat flux was downward at about 20 W m^{-2} (= 2 mW cm^{-2}) required to balance radiational cooling at the surface. The boundary layer was stable, the inversion top being at about 400 metres. Some short time after sunrise the heat flux was reversed in response to incoming solar radiation and a shallow slightly unstable layer formed which began to erode the stable layer. By about 9.30 MST the stable layer was destroyed and the boundary layer deepened more quickly reaching a maximum in early afternoon some 2 or 3 hours after the midday maximum in heat flux. Just before sunset radiational cooling of the ground exceeded solar warming and a stable layer reformed near the ground. Any pollution emitted into the atmosphere during the day now finds itself in a layer which is 'insulated' from the surface and in which the turbulence gradually decays, unable to be replaced since the sources of energy are no longer available. Wind directional shear can now play a much more important role than hitherto since the lack of vertical mixing prevents each molecule of pollution sampling the wind at all levels in the polluted layer. Pollution within the stable layer also undergoes minimal vertical mixing most of the time although it is one of the more interesting characteristics of these stable layers that internal wind gradients increase in response to the removal of shearing stress throwing the balance of forces out of equilibrium, until ultimately the layer becomes dynamically unstable and sudden bursts of turbulence break out for several minutes at a time, bringing fresh pollution close to the ground where dry deposition can play its role. By the end of the night the pollution in the stable layer can have been almost completely removed by this intermittent process. One very clear example of this effect was noted in one of our aircraft sampling-flights described in the next section, where the concentration of SO_2 at about 100 metres was too small to measure after the air had crossed over the cold water of the North Sea (thereby forming a very similar stable layer to that formed over land at night) whereas at greater heights plenty of SO_2 still remained.

In general the amount lost by dry deposition to the ground depends on the concentration just above the ground. The flux is represented as a product of this concentration and a velocity, the so-called velocity of deposition v_d. Since concentration varies with height, v_d strictly must also be height-dependent, although in the first ten metres or so, the variations are relatively small provided v_d is less than a few centimetres per second and the airflow is turbulent. Since the deposition depends on concentration, its magnitude is greatest within the first few kilometres of a low-level source. In the long-range transport problem it is convenient to treat the pollutant as though it were uniformly mixed through the boundary layer (the so-called box-model) and to add an extra initial deposition to allow for the error arising from the fact that the plume from a source takes a finite time to diffuse upwards. During this diffusive process ground-level concentrations

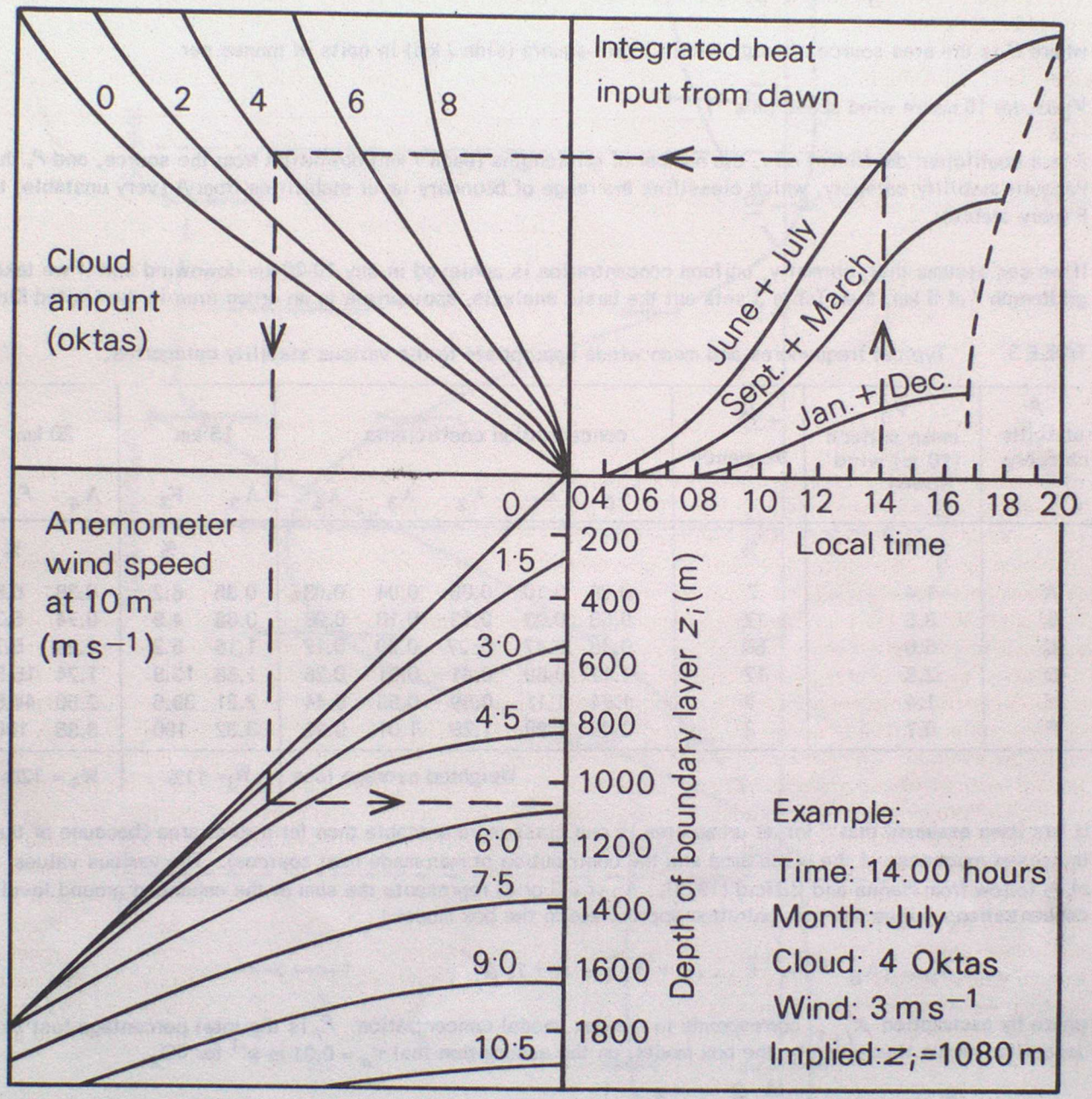

FIGURE 1. A nomogram for estimating the depth of the boundary layer in the absence of marked advective effects or basic changes in weather conditions. The example shows how the diagram is to be used.

and depositions are higher than given by the box-model. Various estimates have been made of this initial deposition and the current OECD model uses for sulphur dioxide a mean value of 15%. This value is supported by very approximate calculations based on the well-known Hanna-Gifford urban pollution model (1973) which says that the concentration c in $\mu g\ m^{-3}$ can be expressed as

$$c = \lambda_i(P)\frac{Q}{V_0\, l^2}$$

where Q is the area source strength within a grid-square (side l km) in units of tonnes per

V_0 is the 10 metre wind speed ($m\ s^{-1}$)

λ is a coefficient dependent on i, the number of gridlengths (each l km) downwind from the source, and P, the Pasquill stability category, which classifies the range of boundary layer stabilities from A (very unstable) to F (very stable).

If we can assume that, normally, uniform concentration is achieved in say 10-20 km downwind and if we take a gridlength l of 5 km, then Table 3 sets out the basic analysis, appropriate to an urban area in the United Kingdom.

TABLE 3 Typical frequencies and mean winds appropriate to the various stability categories.

P stability category	V_0 mean surface (10 m) wind speed	f_P frequency	concentration coefficients					15 km		20 km	
			λ_0	λ_1	λ_2	λ_3	λ_4	Λ_3	F_3	Λ_4	F_4
	$m\ s^{-1}$	%							%		%
A	1.4	7	0.28	0.10	0.05	0.04	0.03	0.35	6.2	0.38	6.8
B	3.5	17	0.53	0.23	0.13	0.10	0.08	0.68	4.9	0.74	5.3
C	5.6	50	0.88	0.47	0.27	0.20	0.17	1.16	5.2	1.28	5.7
D	2.8	17	1.17	0.69	0.41	0.31	0.26	1.56	13.9	1.74	15.5
E	1.4	7	1.64	1.11	0.69	0.53	0.44	2.21	39.5	2.50	44.6
F	0.7	2	2.46	1.98	1.29	1.01	0.85	3.32	100	3.85	100
							Weighted average loss	W_3 = 11%		W_4 = 12%	

It has been assumed that P for an urban area is one class more unstable than for a rural area (because of the increased roughness of the urban area and the contribution of man-made heat sources). The various values of λ_i follow from Hanna and Gifford (1973). Λ_i (i = 3 or 4) represents the sum of the enhanced ground level concentrations minus the concentration appropriate to the box model :

$$\Lambda_i = (\lambda_0 + \lambda_1 + \ldots + \lambda_i) - (i+1)\Lambda_{i+1}$$

where by assumption Λ_{i+1} corresponds to the box model concentration. F_i is the total percentage lost by dry deposition minus that given by the box model, on the assumption that v_d = 0.01 $m\ s^{-1}$ for SO_2.

$$F_i(P) = 0.01 \times \frac{\Lambda_i Q}{V_0} \times l^2 \times \frac{1}{Q} \times 100$$

$$= 25\ \frac{\Lambda_i(P)}{V_0(P)} \quad \text{when } l = 5 \text{ km}$$

Finally the weighted average $W_i \equiv \sum_p \frac{f_P}{100} F_i(P)$

Other values of W_i are : W_5 = 12.89, W_6 = 13.63, W_7 = 14.23, W_8 = 14.95, W_9 = 15.51.

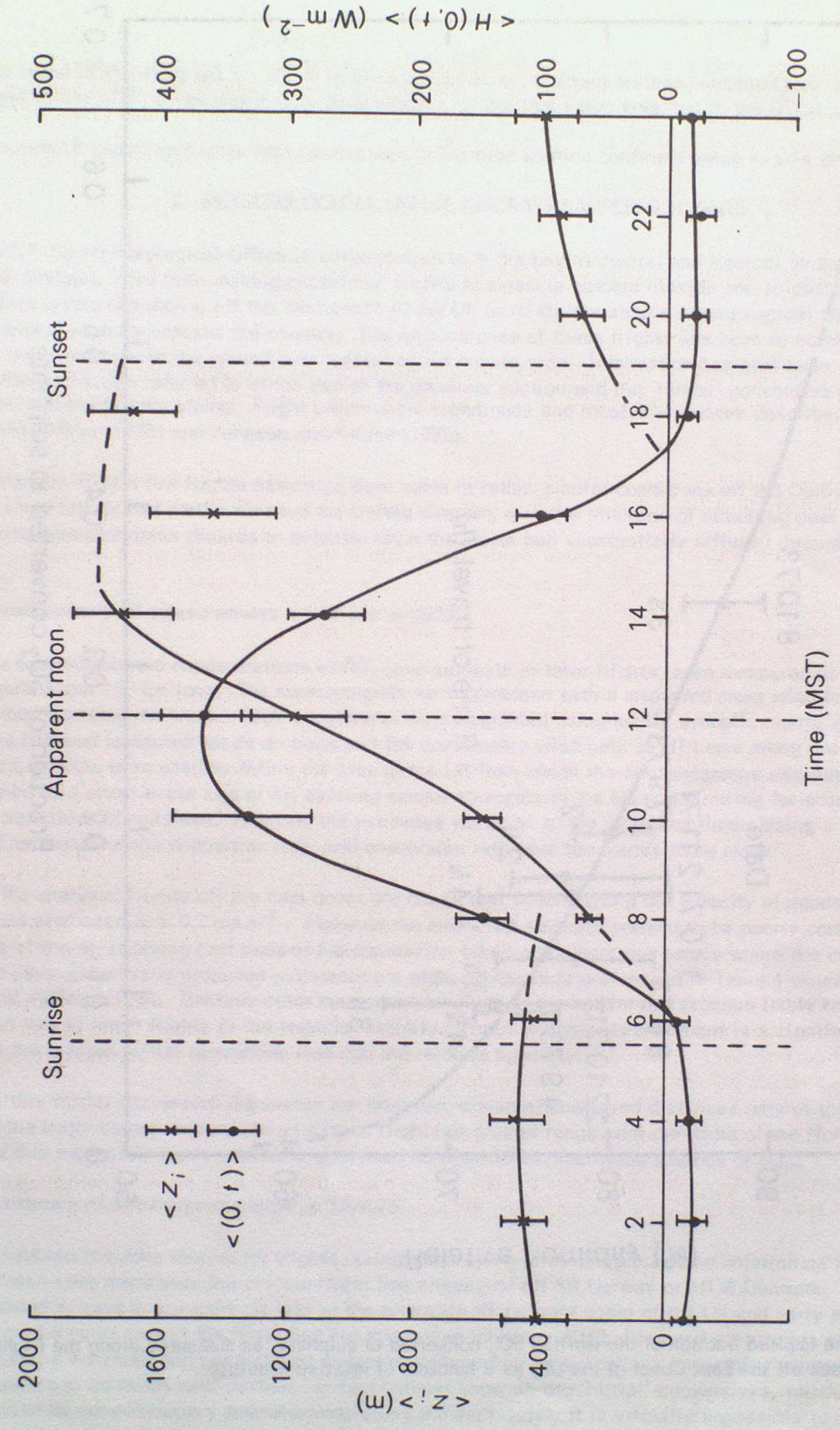

FIGURE 2. The mean boundary layer thickness, $\langle z_i \rangle$, and sensible heat flux at the surface, $\langle H(0,t) \rangle$, deduced for the O'Neill data, plotted with standard errors as functions of time of day, (mean solar time).

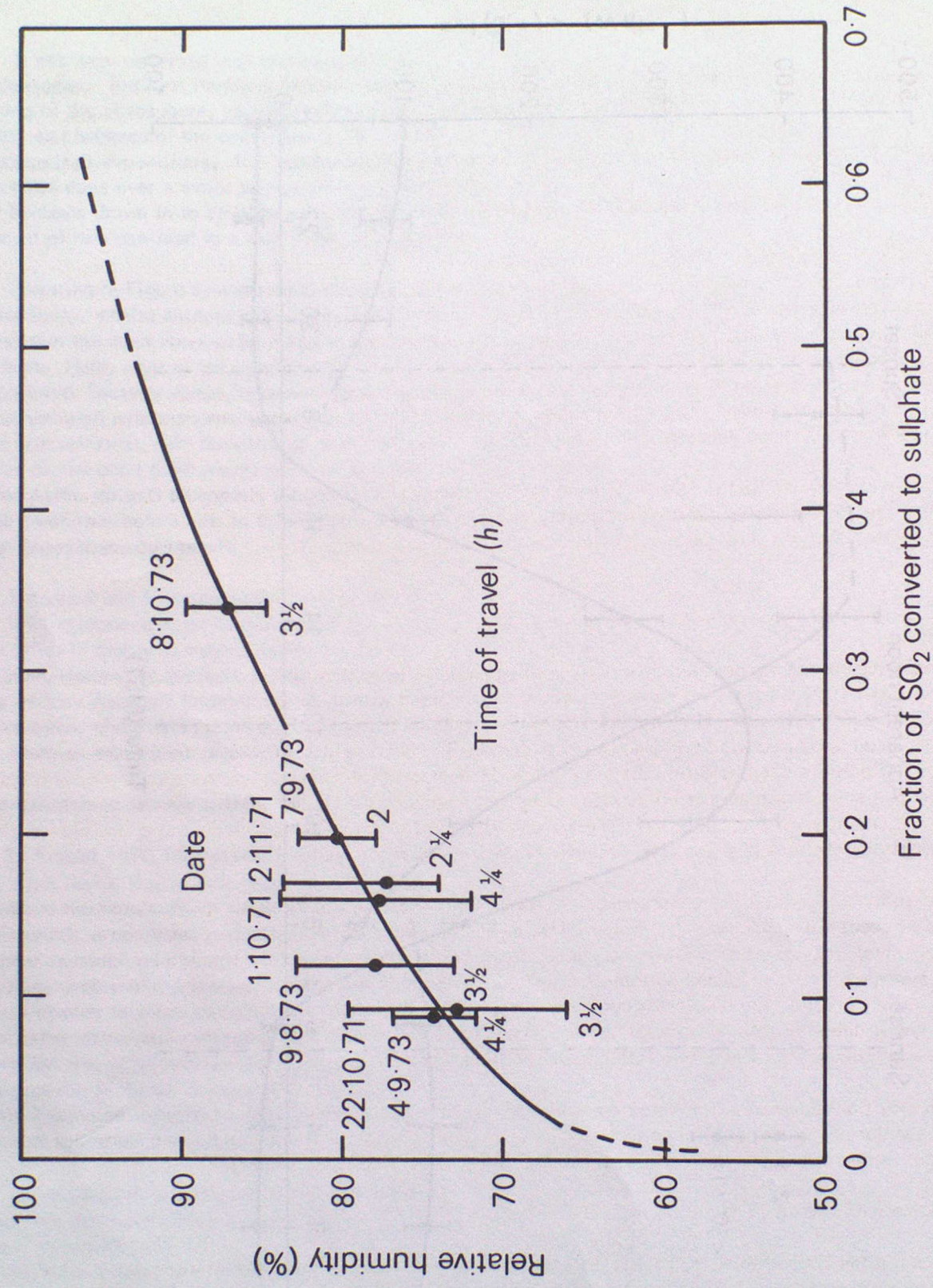

FIGURE 3. The implied fraction of the emitted SO_2 converted to sulphate, as measured along the flight track off the East Coast of the UK, as a function of relative humidity.

The model is therefore not sensitive to the assumption as to where uniform concentration is achieved, and the trend values of W_i gives suprisingly good support for the 15% value selected in the OECD study.

The aircraft sampling flights data summarized in the next section confirm a value of this general magnitude.

2 METEOROLOGICAL OFFICE AIRCRAFT SAMPLING FLIGHTS

Since 1971 the Meteorological Office in collaboration with the Environmental and Medical Sciences Division of AERE, Harwell, have been making occasional flights to measure sulphur dioxide and sulphate aerosol in the mixing layer, and above, off the east coast of the UK in relatively simple meteorological conditions when the air had previously crossed the country. The main purpose of these flights has been to establish the average rate of dry deposition to the ground over typical mixed countryside. Information has also been deduced on the initial deposition referred to at the end of the previous section and the 'initial' conversion of SO_2 to sulphate in the diffusing plume. Flight information, techniques and results have been described in detail in Smith and Jeffrey (1975) and Johnson and Atkins (1975).

Starting in 1974 a few flights have also been made in rather similar conditions off the Danish and Norwegian coasts, when the air had earlier crossed the United Kingdom, with the intention of obtaining data on the conversion rate of sulphur dioxide to sulphate once the plume had substantially diffused through the mixing layer.

2.1 *Brief summary of measurements to the end of 1973.*

In these experiments the concentrations of SO_2 (and sulphate in later flights) were measured at several levels over a path about 150 km long. The measurements were combined with a measured mean wind to give the flux of pollutant crossing the track. Back trajectories were estimated using hourly synoptic charts assuming the relations between measured winds on track and the geostrophic wind held at all times along the trajectory. These trajectories were used to define the area of the UK from which the SO_2 emissions originated, which were determined by a small-scale map of all emitting source strengths in the UK and allowing for seasonal and diurnal variations of emission. Relating the estimated emission to the measured fluxes using a simple model enabled estimates of the deposition rate, and conversion rate near the source to be made.

All the analysed flights off the east coast are consistent in estimating the velocity of deposition of SO_2 to the ground at about 0.8 ± 0.2 cm s^{-1}. However, the measured sulphate seemed to be poorly correlated with the distance of travel, implying that most of the conversion takes place near the source where the concentrations of SO_2 and other industrially produced pollutants are high. These data are set out in Table 4 which is taken from Smith and Jeffrey (1975). The only other component involved in the conversion process likely to vary in an important way in these flights is the relative humidity. Figure 3 suggests that there is a significant relationship between the implied initial conversion rate and the relative humidity.

That this initial conversion dominated the total conversion at downwind distances only of the order 100-200 km from the major source region, led us to plan flights at greater range, and the width of the North Sea seemed ideal for this especially since it clearly contained no significant man-made sources of SO_2.

2.2 *A summary of measurements made in 1974-75*

Table 5 sets out the main results for flights during these two years, using the same notation as in Table 4. Five of these were made over the eastern North Sea either just off SW Norway or off W Denmark. Originally it was intended to sample the same air late in the afternoon off the east coast of the UK and early the next morning on the other side of the North Sea but this posed too many difficulties and constraints on the meteorology, the aircraft logistics and on trajectory forecasting. Implications concerning conversion rates have therefore to be taken with caution. In fact without knowing the 'initial' conversions, which could have been implied by complementary measurements along the east coast, it is virtually impossible to deduce this rate. Only in one flight on 22 August 1974 can an estimate be made. On this occasion there were two layers; a lower layer with an inversion extending up to about 260 metres in which nearly all the SO_2 had been deposited or converted to sulphate, and an upper layer in which the loss of SO_2 was assumed to be due to conversion to sulphate only.

TABLE 4 THE SULPHUR BUDGET

Quantity	Symbol	Meaning	Unit	Date						
				1.10.71	22.10.71	2.11.71	9.8.73	4.9.73	7.9.73	8.10.73
	z	depth of mixing layer	m	1050	450	600	1200	1700	1200	1600
	V	mean speed of sampled air	m s^{-1}	10.0	20.5	17.3	14.7	8.1	12.5	6.9
	x	distance from major upwind source	km	100	220	140	190	125	87	90
	t	time of travel from major source	h	4.25	3.5	2.25	3.5	4.25	2	3.5
	σ_z	r.m.s. vertical extent of SO_2 plume on sampling track	m	600	260	346	580	760	280	540
	σ_z (neut.)	theoretical value for neutral atmosphere	m	680	1100	800	900	750	640	645
	r	indicator of significant rain		—	—	—	—	—	—	r
Emissions	E_2	estimated emission flux of SO_2 from sources	t h^{-1}	238	158	370	149	320	67	78
Sulphur dioxide	M_2	measured SO_2 flux on sampling track	t h^{-1}	122	112	193	85	187	32	27
	M_2/E_2	fraction of emitted SO_2 remaining off the east coast		0.51	0.71	0.52	0.57	0.58	0.48	0.34
	v_d	deduced velocity of dry deposition	cm s^{-1}	—	—	—	0.8	0.8	0.6	—
		assumed v_d implied by comparison	cm s^{-1}	0.8	0.8	0.8	—	—	—	0.7
	D_2/E_2	fraction of emitted SO_2 lost by dry deposition		0.33	0.20	0.31	0.31	0.33	0.32	0.32
	C_2	average SO_2 concentration on sampling track	μg m^{-3}	30	15	25	10	22	6	7
Sulphate (converted to equivalent SO_2)	M_4	measured sulphate flux on sampling track	t h^{-1}	—	—	—	18	30	13	6
		deduced sulphate flux on sampling track	t h^{-1}	38	14	63	—	—	—	—
	M_4/E_2	fraction of emitted SO_2 appearing as sulphate on track		0.16	0.09	0.17	0.12	0.09	0.20	0.08
	R_4/E_2	R_4 = amount of sulphate removed by rain		0	0	0	0	0	0	0.26
	R_4/P_4	P_4 = total production of sulphate from SO_2		0	0	0	0	0	0	0.75
	P_4/E_2	($P_4 \equiv M_4 + R_4$)		0.16	0.09	0.17	0.12	0.09	0.20	0.34
Relative humidity	—	mean relative humidity within plume	%	78	73	78	78	74	81	87
	—	range about mean	%	±6	±7	±4	±5	±3	±3	±3
	P_4^*/E_2	P_4^* = production of sulphate implied by empirical curve (Figure 3)		0.15	0.08	0.15	0.15	0.09	0.21	0.33
Total Budget		$100\ (M_2 + D_2 + P_4)\ /E_2 \equiv 100$		100	100	100	100	100	100	100
		$100\ (M_2 + D_2 + P_4^*)\ /E_2$		99	99	98	103	100	101	99

Notes : (a) all fluxes are measured in tonnes per hour.

(b) the basic known emission and flux data are E_2, M_2 and (in 1973 only) M_4. All other items are deduced from budget requirements and (in 1971 and on 8.10.73) by taking the assumed value of v_d.

(c) on all occasions except 22.10.71 the SO_2 plume had not completely filled the mixing layer and the loss by dry deposition to the ground was deduced using a developing plume model (see text). On 22.10.71 the shallow mixing layer was completely filled and a simple 'box-model' was used instead.

TABLE 5 RESULTS OF SEVEN FLIGHTS IN 1974-75

Date	E_2 estimated emission	M_2 flux of SO_2	M_2/E_2 %age of SO_2 remaining	M_4 flux of SO_4	M_4/E_2 %age of SO_2 as SO_4 remaining	D_2/E_2 %age of E_2 deposited	z_i mixing depth	V mean speed	x distance from source	t time from source	Remarks
	$t\ h^{-1}$	$t\ h^{-1}$	%	$t\ h^{-1}$	%	%	m	$m\ s^{-1}$	km	h	
21.5.74	35.4	8.3	23.4	14.7	41.5	35.1	2000	9.3	650	23	Track off W. Denmark. Air accelerated over North Sea
22.8.74 (above inversion)	–	conc.SO_2 = 16.3 $\mu g\ m^{-3}$	–	conc.SO_4 = 7.3 $\mu g\ m^{-3}$	–	–	2700 (height of inversion)	7.2	700	18	Track off S.W. Norway. On assumption that concentrations of SO_2 were identical above and below.
(within surface inversion	–	= 1.4 $\mu g\ m^{-3}$	–	= 5.3 $\mu g\ m^{-3}$	–	–	top ≈ 260 m	7.2	700	18	260 m off the UK east coast, then the measured values imply an $SO_2 \rightarrow SO_4$ conversion of 1% per hour.
28.8.74	323	131	40.5	36	11.1	48.4	1700	9.3	150	4.5	Track off UK east coast. A rather high apparent loss D_2/E_2
20.12.74	173	53	30.4	25	14.5	55	1600	23	460	5.5	Track over eastern part of North Sea
5.2.75	515	244	47.4	60	11.7	40.9	900	13	over Europe		Track west of Cornwall. Trajectory back-tracked to N. Italy where precipitation was falling.
20.2.75	534	≈ 3	–	≈ 5	–	–	500	12	700	>14	Track off SW Norway. Hand & computer back-tracks crossed major sources in UK. However z was low and track strongly affected by surface inversion over UK the previous night. Low values confirmed by Norwegian surface data.
26.2.75	–	–	–	–	–	–	–	–	–	–	An example of the very occasional run which yielded inconsistent and poor quality analytical data, largely due to a very poor batch of filter paper.

Assuming that both layers had identical SO_2 and sulphate concentrations on leaving the east coast 18 hours earlier, time constants for the exponential decay of SO_2 by deposition (7.3 hours) and conversion (99 hours) could be deduced. The latter is equivalent to a conversion rate of 1% per hour.

Such a slow rate reflects back on the whole concept of double sampling the same air on both sides of the North Sea. The limits of precision of the aircraft system for measuring SO_2 and sulphate are such that a conversion rate as low as 1% per hour would be detected with difficulty and would be subject to much uncertainty. There seems little hope therefore of establishing a relationship between this rate and other atmospheric variables such as relative humidity with our present sampling system.

It is interesting that 1% per hour agrees very well with other estimates. Eliassen and Saltbones (1975) have analysed ground-level measurements in terms of a simple model in which the SO_2 concentrations are subject to a first order decay due to deposition and conversion. They found decay rates varying between 1.7% per hour and 0.3% per hour. Recently Lusis and Wiebe (1976) in a study of the INCO nickel smelter-stack plume at Sudbury in Canada, which emits an average of 3500 tonnes SO_2 per day, deduced an average 1% per hour conversion rate. They failed to find any dependence on temperature or relative humidity.

Returning to Table 5, one flight is clearly anomalous, namely that of 20 February 1975 when very small concentrations were recorded. Nearly all the apparent emission had been lost, and whether this can be explained by a widespread and fairly deep nocturnal surface inversion over England during the passage of the air that was later sampled, is a matter of conjecture, (see Figure 4). At least the small measured concentrations appear to be genuine since they are consistent with very low measurements recorded at ground level in SW Norway.

Flights on 21 May, 28 August, 20 December and 5 February provided complete and useful data. Being in general at longer range from the major sources upwind than those in Table 4, it is consistent that the estimated percentage losses by deposition are that much greater. Subtracting the average estimated 'initial' loss of 15% from both sets of losses and taking average fluxes and distances of travel, the figures imply a velocity of deposition over land of 0.8 cm s^{-1} and very approximately 0.5 cm s^{-1} over the sea.

2.3 *Advecting winds*

One of the more important problems in attempting the determination of back trajectories is to relate the 'advecting wind', that is the mean wind affecting the pollution in the mixing layer at every stage along the track, to some known wind such as the surface geostrophic wind. Any such relationship is clearly going to be subject to error, especially if the track runs into an area of strong curvature near the centre of a depression or if the air mass is strongly baroclinic. In maritime areas such as western Europe the errors are normally acceptable as is evidenced by the strong similarity between trajectories based on surface geostrophic winds and those using 850 mb winds (see the OECD LRTAP Report). On the other hand, over continental regions in winter, say over Canada or the USSR, it is very apparent that the strong stability of the lower layers of the atmosphere renders the use of the surface geostrophic wind inappropriate and an 850 mb wind approach is much better.

The great advantage of using the surface geostrophic wind where it is justified is that surface pressure charts are usually available at one-hourly intervals whereas winds at other pressure levels are only at six-hourly intervals, and this difference must affect the relative accuracies of the trajectories.

In the aircraft sampling experiments, real winds were measured by the aircraft and were related to the local surface geostrophic winds and to the surface 10-m winds when these were available. The following relatively simple procedure was used; a mean wind normal to the track was formed from the measured winds weighting the value at each level by the measured concentration to get a mean wind for the sulphur dioxide plume. The resulting speed V_p was compared with the local surface geostrophic wind speed V_G and the ratio V_p/V_G was assumed constant along the back trajectory. The mean plume-wind direction θ_P on the other hand was compared with both the geostrophic wind direction θ_G and the surface (10-m) wind direction θ_0 and the ratio $(\theta_G - \theta_P)/(\theta_G - \theta_0)$ was also assumed constant along the back trajectory. For the 1971-73 flights the

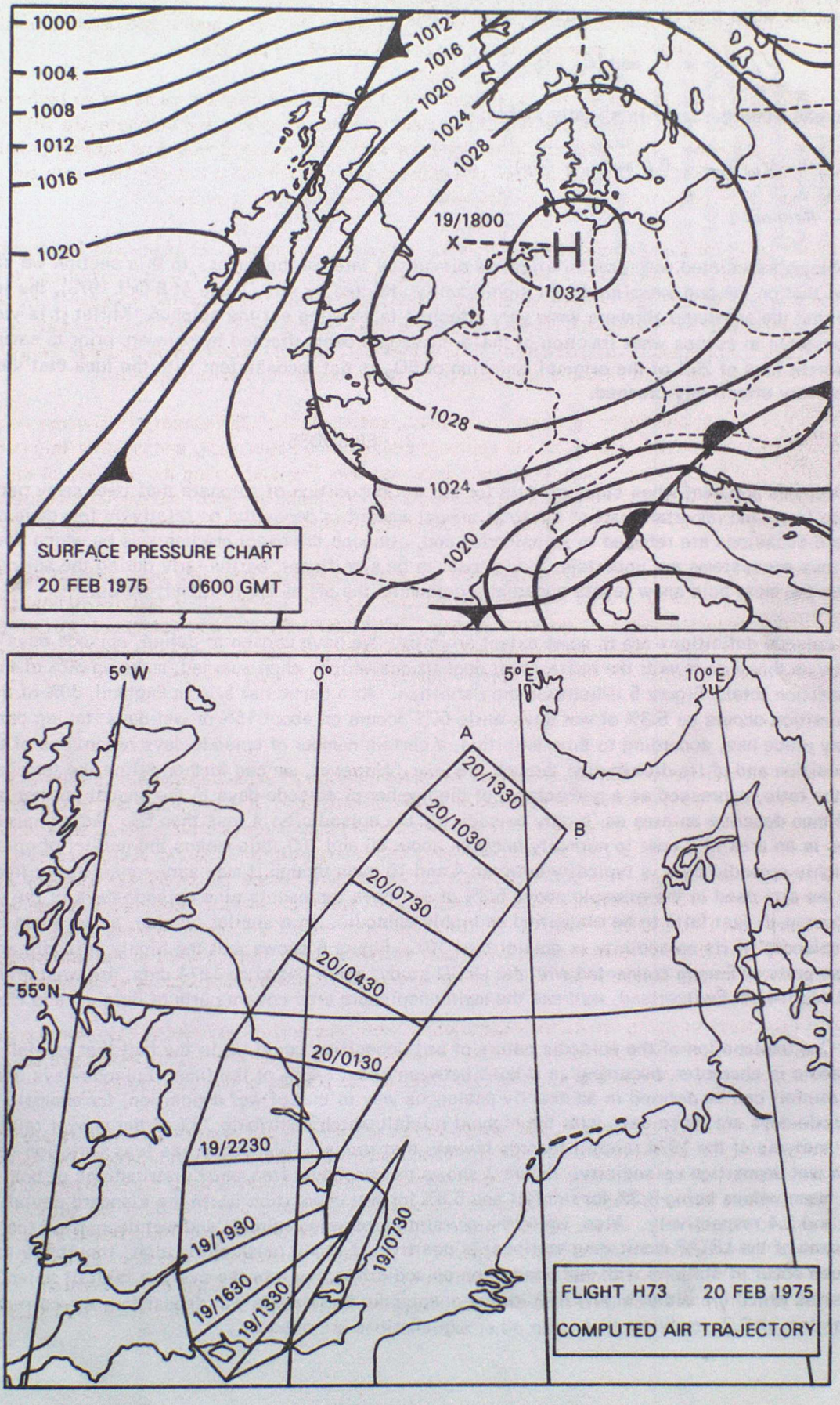

FIGURE 4. The synoptic situation and computed back-trajectory for the sampling flight on 20 February 1975

ratio V_P/V_G and the angular difference $(\theta_G - \theta_P)$ are both well related to the overall stability of the boundary layer, for when this is near neutral or slightly unstable:

$$V_P/V_G \approx 1 \quad \text{and} \quad (\theta_G - \theta_P) \approx 10°$$

whereas when the layer is slightly stable:

$$V_P/V_G \approx 0.9 \quad \text{and} \quad (\theta_G - \theta_P) \approx 20°$$

2.4 *Rain-out*

Problems associated with precipitation are discussed later in the paper. In this section we simply wish to note that on the one sampling flight significantly affected by rain (Table 4: 8 Oct.1973), the implications are that the scattered showers were very effective in washing out the sulphur. Whilst it is virtually impossible to assess what fraction of the airmass had been affected by showers prior to sampling, the apparent loss of 26% of the original emission of SO_2 is not inconsistent with the idea that those areas had been very effectively cleaned.

3. EPISODES

It becomes apparent when studying data for the wet deposition of sulphate that over many parts of Europe a quite large and important part of the total annual amount is deposited on relatively few days of the year. These occasions are referred to as episodes and, although the exact mechanisms by which they affect the various ecosystems are uncertain, they appear to be significant, particularly during the spring snow melt when the most acid snow layers apparently determine the pH of the first melt-water.

Episode definitions are to some extent arbitrary. We have chosen to define 'episode-days' at a particular place as those days with the highest wet depositions which, when summed, make up 30% of the annual wet deposition total. Figure 5 illustrates the definition. At a particular site in England, 30% of the wet sulphate deposition occurs on 5.3% of wet days while 50% occurs on about 15% of wet days (taking data for 1974). Every place has, according to this definition, a certain number of episode-days regardless of the total deposition and of its distribution through the year. However, we can further define the term episodicity' as the ratio, expressed as a percentage, of the number of episode-days to the annual number of wet days and then describe an area as 'highly episodic' if the episodicity is less than 5%. As the number of wet days in an area in a year is normally between about 80 and 200, this means the number of episode-days in a highly episodic area is typically between 4 and 10 even though it may vary considerably from year to year. For the site used in the example above 5.3% of wet days represents nine episode-days in the year and, of course, it just fails to be classified as highly episodic. In a similar fashion, an area can be classified 'unepisodic' if its episodicity is greater than 10%. Figure 6 shows that the highly episodic areas in those parts of Europe connected with the OECD study, again based on 1974 data, are western Norway, and south-west Switzerland, whereas the main unepisodic area covers parts of Belgium and Holland.

One explanation of the episodic nature of wet deposition could lie in the fact that rainfall itself is episodic in character, occurring as it does between about 5-20% of the time. Episode-days and episodicity for rainfall can be defined in an exactly analogous way to that of wet deposition; for example rainfall episode-days are those days with the highest rainfall which contribute 30% of the annual rainfall, but analysis of the 1974 rainfall records reveals that rainfall episodicity has less variation geographically than wet deposition episodicity. Figure 7 shows the smoothed frequency distributions of both quantities, the mean values being 8.3% for rainfall and 6.9% for wet deposition while the standard deviations are 1.7 and 2.4 respectively. Also, while the correlation between rainfall and wet deposition (calculated at some of the LRTAP monitoring stations) is positive, it is not particularly high. Inevitably the highest values occur at stations with wet deposition episodicities close to the average rainfall episodicity. At sites which are either highly episodic or unepisodic in sulphate wet deposition, the correlation falls to as low as 0.2, implying that some other explanations are needed.

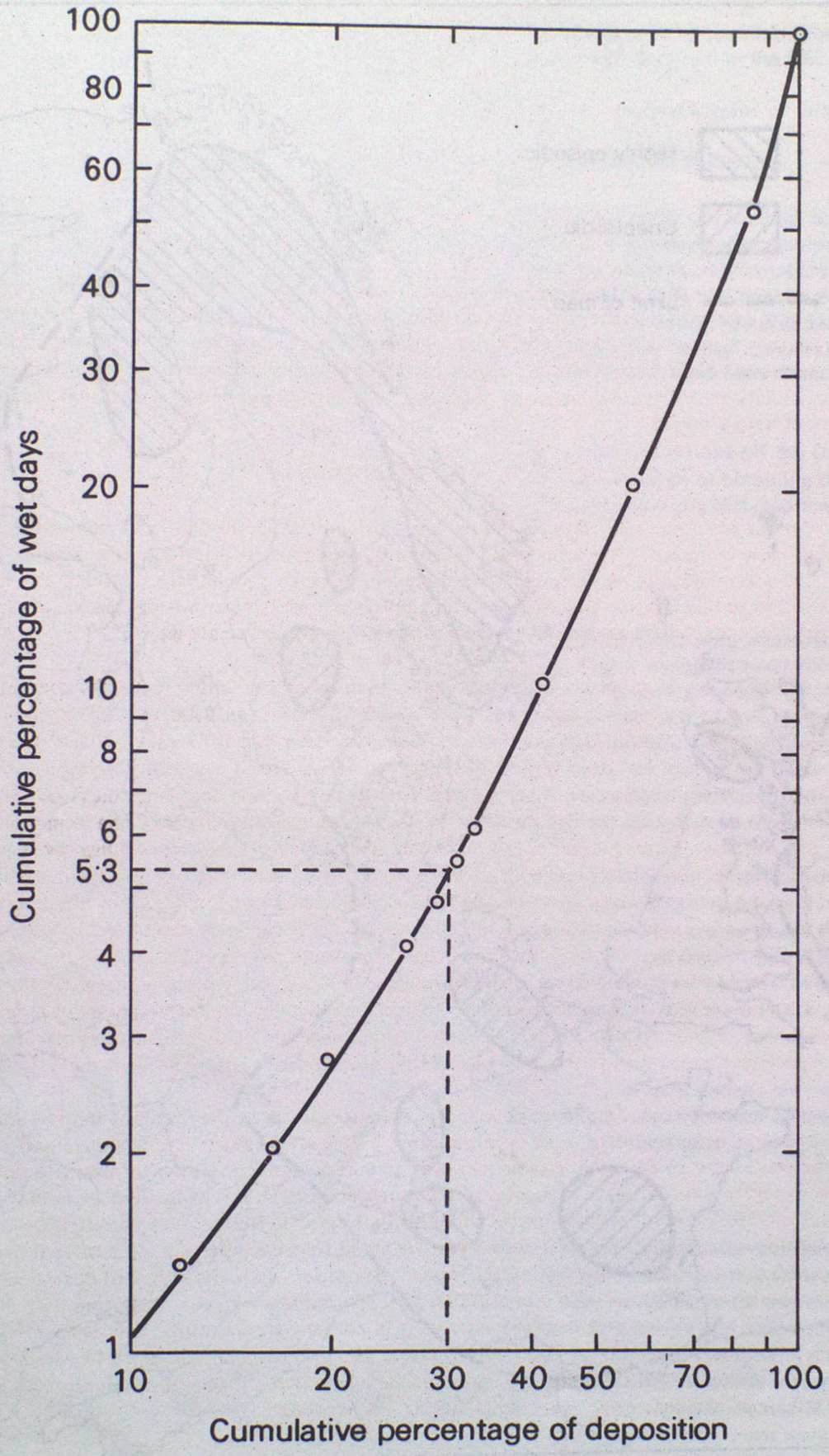

FIGURE 5. The cumulative percentage of deposition plotted against the cumulative percentage of wet days, plotted on logarithmic scales, at Cottered

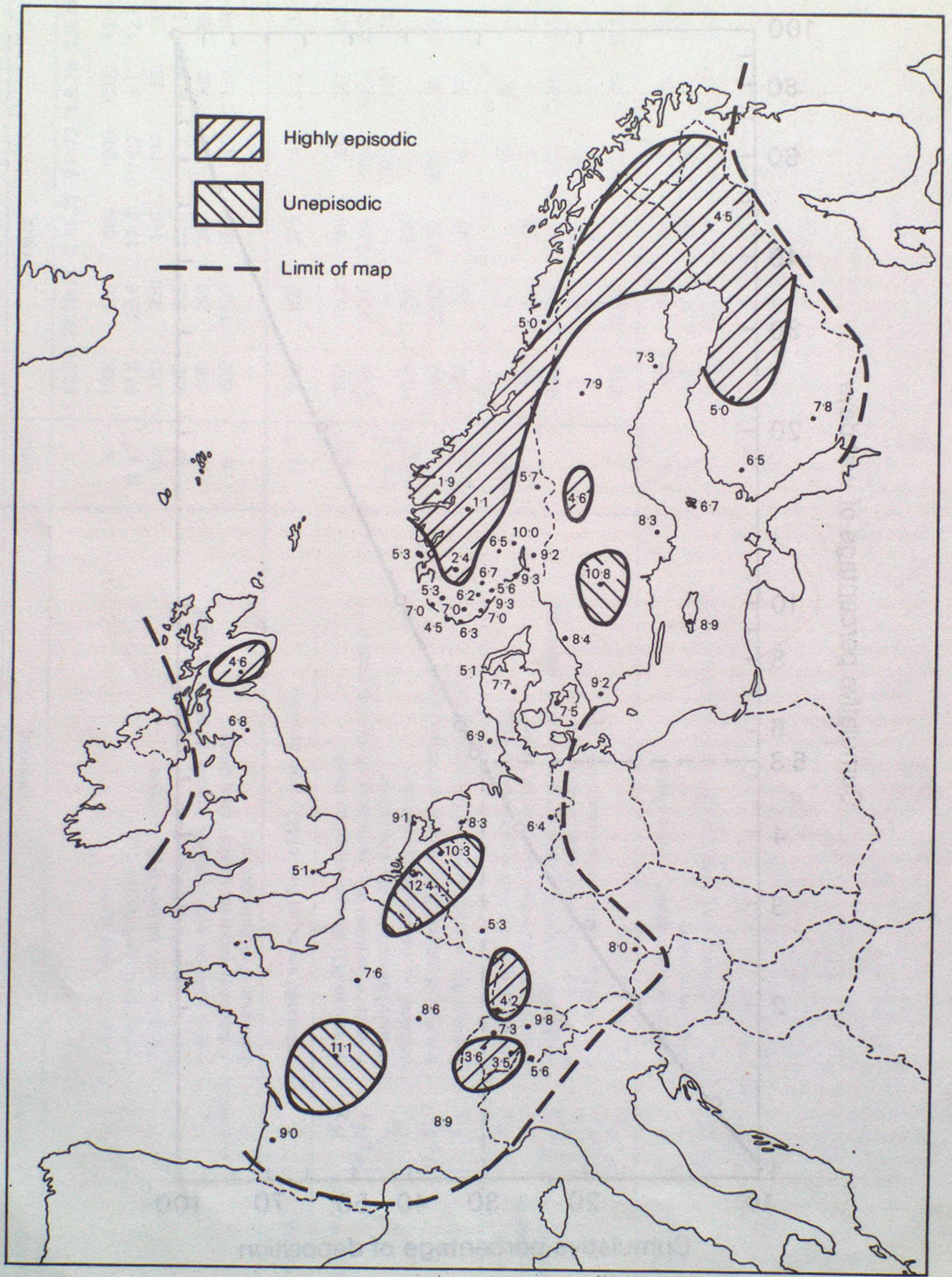

FIGURE 6. A map of Western Europe showing the distribution of episodicity for 1974

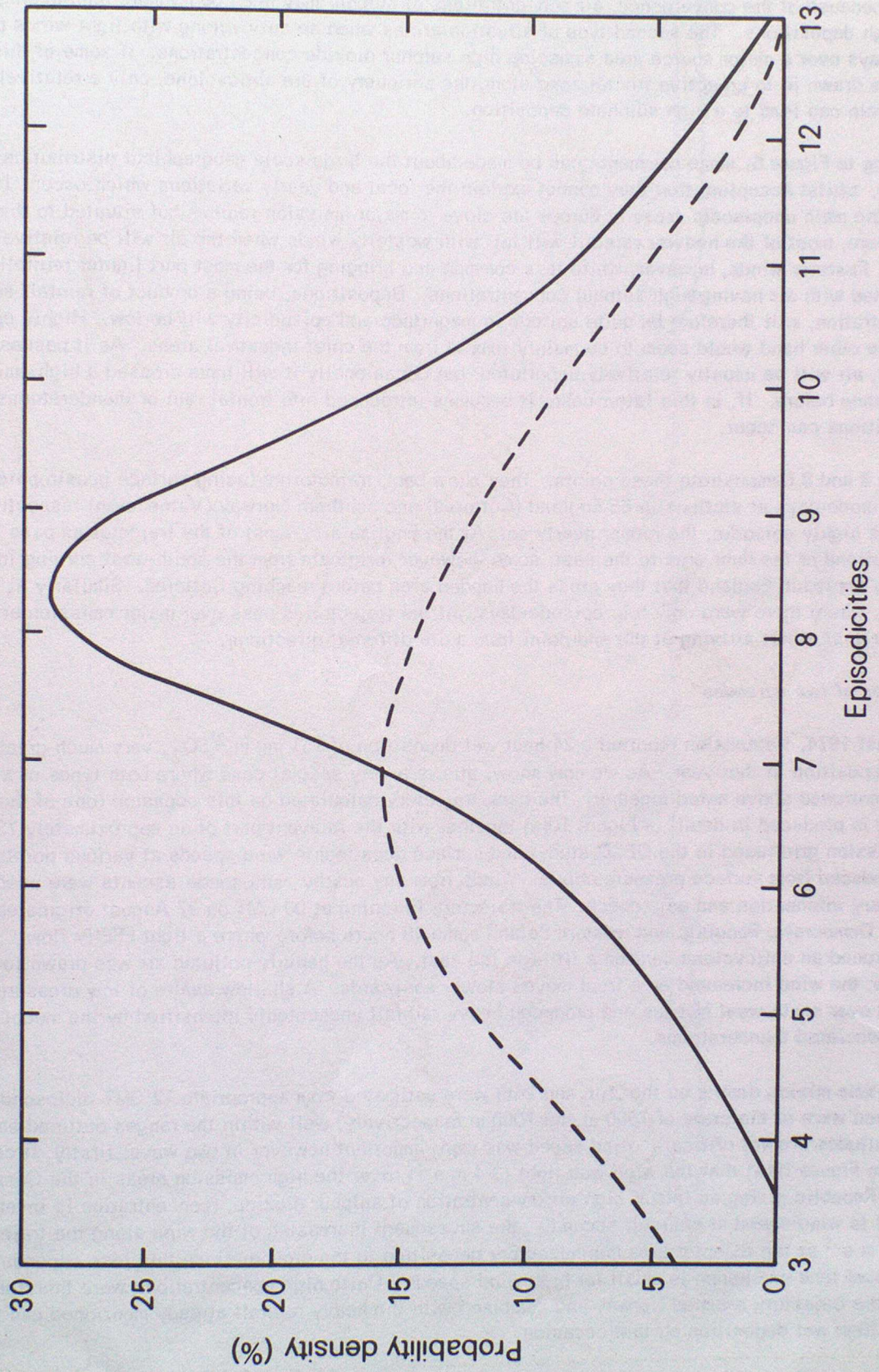

FIGURE 7.₁ Episodicities for rainfall (full line) and sulphate deposition (packed line)

It has been observed that two main types of synoptic situation seem to be associated with the occurrence of episodes. The first involves slow-moving active frontal systems where convergence of moist air at low levels of the atmosphere, usually enhanced by mountain ranges, produce heavy and prolonged rainfall. In this case, and because of the convergence, air concentrations of sulphur may need be no more than average to produce high depositions. The second type of situation arises when an anticyclone with light winds persists for a few days over a major source area amassing high sulphur dioxide concentrations. If some of this polluted air becomes drawn in to an active frontal zone along the periphery of the anticyclone, only a relatively small amount of rain can lead to a high sulphate deposition.

Referring to Figure 6, some comments can be made about the large-scale geographical distribution of episodicity, whilst accepting that they cannot explain the local and yearly variations which occur. It would seem that the main unepisodic areas in Europe are close to major emission regions but situated to the west of them. Here, most of the heavier rainfall will fall with westerly winds when the air will be relatively unpolluted. Easterly winds, however, while less common and bringing for the most part lighter rainfalls, will be associated with air having high sulphur concentrations. Depositions, being a product of rainfall amount and concentration, will therefore be quite uniform in magnitude and episodicity will be low. Highly episodic areas on the other hand would seem to be mainly remote from the chief industrial areas. As it passes over these parts, air will be usually relatively unpolluted, but occasionally it will have crossed a high emission area some time before. If, in this latter case, it becomes introduced into frontal rain or thunderstorms very high depositions can occur.

Figures 8 and 9 demonstrate these points. They show back-trajectories (using surface geostrophic winds) for 1974 episode-days at stations in SE England (Cottered) and southern Norway (Vatnedalen) respectively. The latter is highly episodic, the former nearly so. At the English site, most of the trajectories pass over Belgium, Holland or the Ruhr area to the east; some, however, originate from the south-west curving in such a way as they approach England that they cross the London area before reaching Cottered. Similarly at Vatnedalen, where there were only four episode-days, all the trajectories pass over major emission areas to the south or east albeit arriving at the end-point from quite different directions.

Case studies of two episodes

On 26 August 1974, Vatnedalen reported a 24-hour wet deposition of 151 mg m^{-2} SO_2, very much greater than any other deposition in that year. As we now show, this is a very special case where both types of synoptic situation mentioned above acted together. The back-trajectory calculated on this occasion (one of those shown in Figure 9) is produced in detail in Figure 10(a) together with the relevant part of an approximately 127 x 127 km sulphur emission grid (used in the OECD study) and surface geostrophic wind speeds at various points along the route deduced from surface pressure charts. Winds from any nearby radiosonde ascents were used as supplementary information and as a check. The trajectory finishing at 00 GMT on 27 August originates from the German Democratic Republic and western Poland some 48 hours before where a light ESE'ly flow prevailed around an anticyclone centred a little to the east. As the heavily polluted air was drawn towards Scandinavia, the wind increased as a front moved slowly eastwards. A shallow centre of low pressure formed on the front over south-west Norway and produced heavy rainfall undoubtedly intensified by the mountains and with associated thunderstorms.

The daytime mixing depths on the 25th and 26th were estimated from appropriate 12 GMT radiosonde soundings and were of the order of 1500 m and 1000 m respectively, well within the ranges outlined earlier (exact magnitudes are not critical). Wind speed was more important however in two ways: firstly, it can be seen from Figure 10(a) that the wind was light (3-4 m s^{-1}), over the high emission areas in the German Democratic Republic giving an initial high air concentration of sulphur dioxide, (concentration is inversely proportional to wind speed at source); secondly, the subsequent increasing of the wind along the trajectory to reach 14 m s^{-1} at the receptor area minimized dry deposition to the ground, since this loss depends on the total travel time and hence is small for high wind speeds. Quite high concentrations were therefore still present as the trajectory reached Norway and combined with the heavy rainfall already mentioned can then explain the high wet deposition on that occasion.

The second example for consideration is that of 7 August 1974 at Cottered. Figure 10(b) shows the calculated back-trajectory finishing at 00 GMT on the 8th. The wet deposition of 119 mg m^{-2} SO_2 was significantly greater than for any other occasion that year. The synoptic situation over the days leading to the episode has similarities with the first example, the trajectory originating from an anticyclonic region with light winds. On this occasion, an area of high pressure close to south-east England on the 6th drifted slowly east and declined allowing a SE'ly flow to develop over the Low Countries. Meanwhile a trough of low pressure was moving into France and as this approached south-east England late on the 7th, outbreaks of rain and thunderstorms started to occur lasting for several hours. Wind speeds along the trajectory ranged from 3 m s^{-1} over the Federal Republic of Germany to 13-14 m s^{-1} over Belgium reducing to near 10 m s^{-1} at the receptor area. Again as with the previous example, the mixing depths at 12 GMT on the two days prior to the episode back along the trajectory were unremarkable - about 1500 m on the first day and 1100 m on the second. Major emission areas in Belgium were traversed by the trajectory but, contrary to the first example, the wind speed here was quite high. The lighter winds earlier were over only moderate emission areas. Clearly then the arguments used to account for the deposition in the first case are only partly effective in the second.

Applying the statistical trajectory model in its general form predicted a wet deposition of 85 mg m^{-2} compared with an observed 119 mg m^{-2}. However, correcting the model for the real observed winds, for probable diurnal variations in emissions, for the observed mixing depths and the duration of rainfall at Cottered, reduces the predicted deposition to 59 mg m^{-2}, roughly half the observed value. One probable explanation for this discrepancy is that the individual storm cells represent active centres of convergence into which polluted boundary layer air can continually be drawn and from which the sulphate can be removed by washout. This convergence, necessary for the maintenance of the storm, can produce up-draught regions 1-2 km across in the lower parts of the cloud with vertical velocities frequently exceeding 1 m s^{-1} and often 10 m s^{-1}, implying normal local horizontal convergence rates of 10^{-3} to 10^{-2} s^{-1}.

On this occasion the cells were moving slowly in a NW direction. Without convergence the sulphate within and below the cloud would rapidly be removed, and by the time Cottered was reached very little deposition would be expected. In order to give twice the model-predicted deposition it is necessary that during the passage of an individual cell an amount of air equalling twice the mixing-depth volume below the cloud would have to be drawn into the cell from surrounding areas. If the radius of the active cell is r (metres) then the convergence C is directly related to the inflow of air:

$C \pi r^2 z$ = inflow around the boundary within the mixing layer depth z, per second.
= 2 x (volume of this sub cloud air)/(time of passage of cloud)

$$= 2\pi r^2 z \times \frac{V}{2r} = \pi r z V$$

i.e. $C = V/r$

On this occasion $V \approx 3$ m s^{-1}, and taking r as being very roughly 10^3 m (1 km)
then: $C \approx 3 \times 10^{-3}$ s^{-1}, falling well within the expected range.

When considering episodes in this manner, it should be noted that errors in calculating trajectories assume a much greater importance than when considering, say, annual statistics of pollution budgeting. Only small errors in the direction of the wind near to the receptor area can and often do make a large difference in determining which regions have been crossed a day or two beforehand. In fact on the occasion of the first example trajectories calculated from the 850 mb wind, at a level strictly above the mixing layer, (OECD, 1977) indicate the air having come from as far west as the English Channel area. It should also be recalled that vertical mixing in the polluted layer is assumed sufficiently good for the plume to move with a uniform vector velocity and wind shear effects are fairly minimal. The importance of this latter effect is considered below.

Although these two episodes were chosen for further study because they had the highest depositions at their sites in 1974, and are therefore in that sense extreme, they nevertheless show how the various

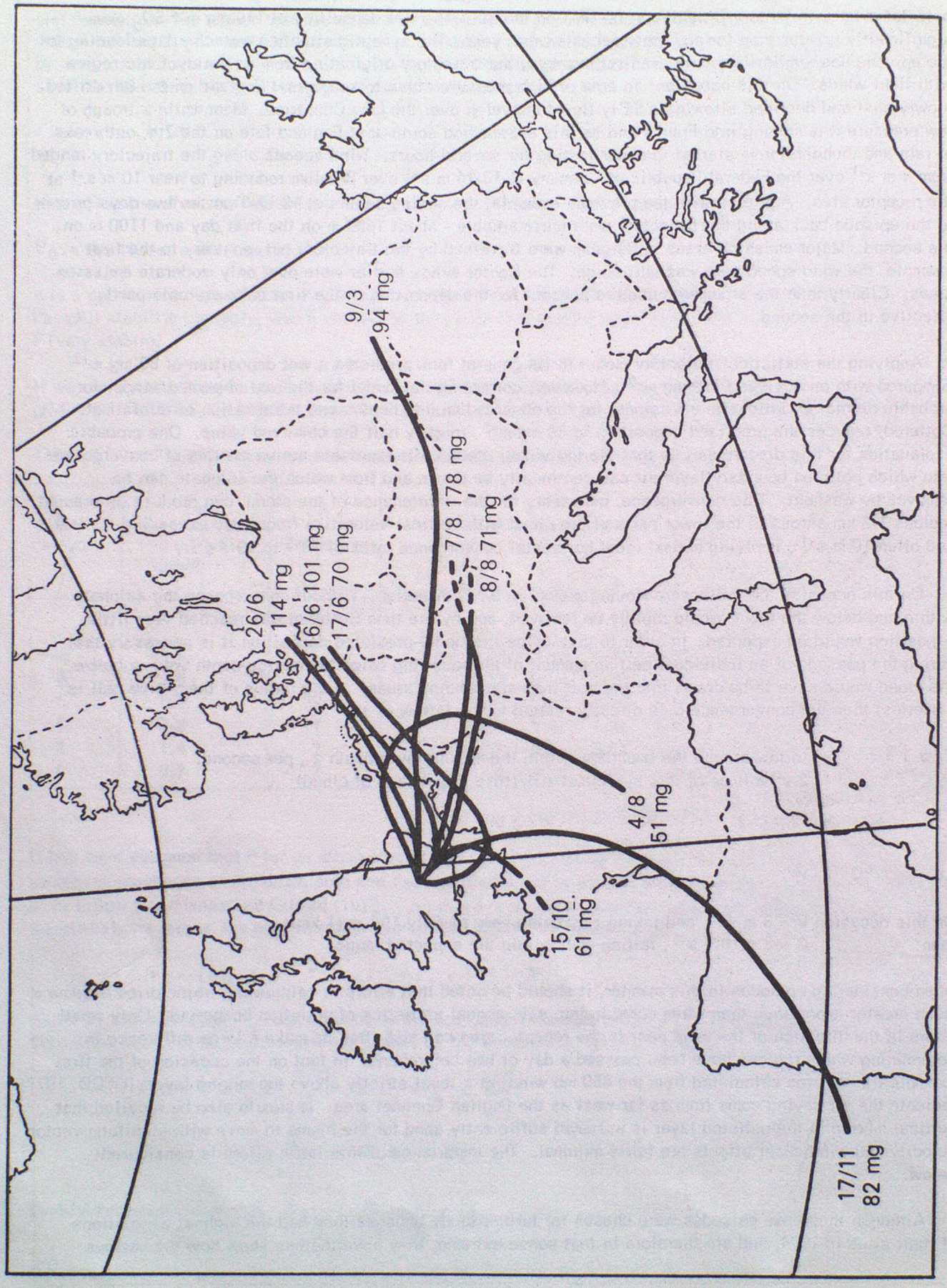

FIGURE 8. Back trajectories for the episode days at Cottered, 1974

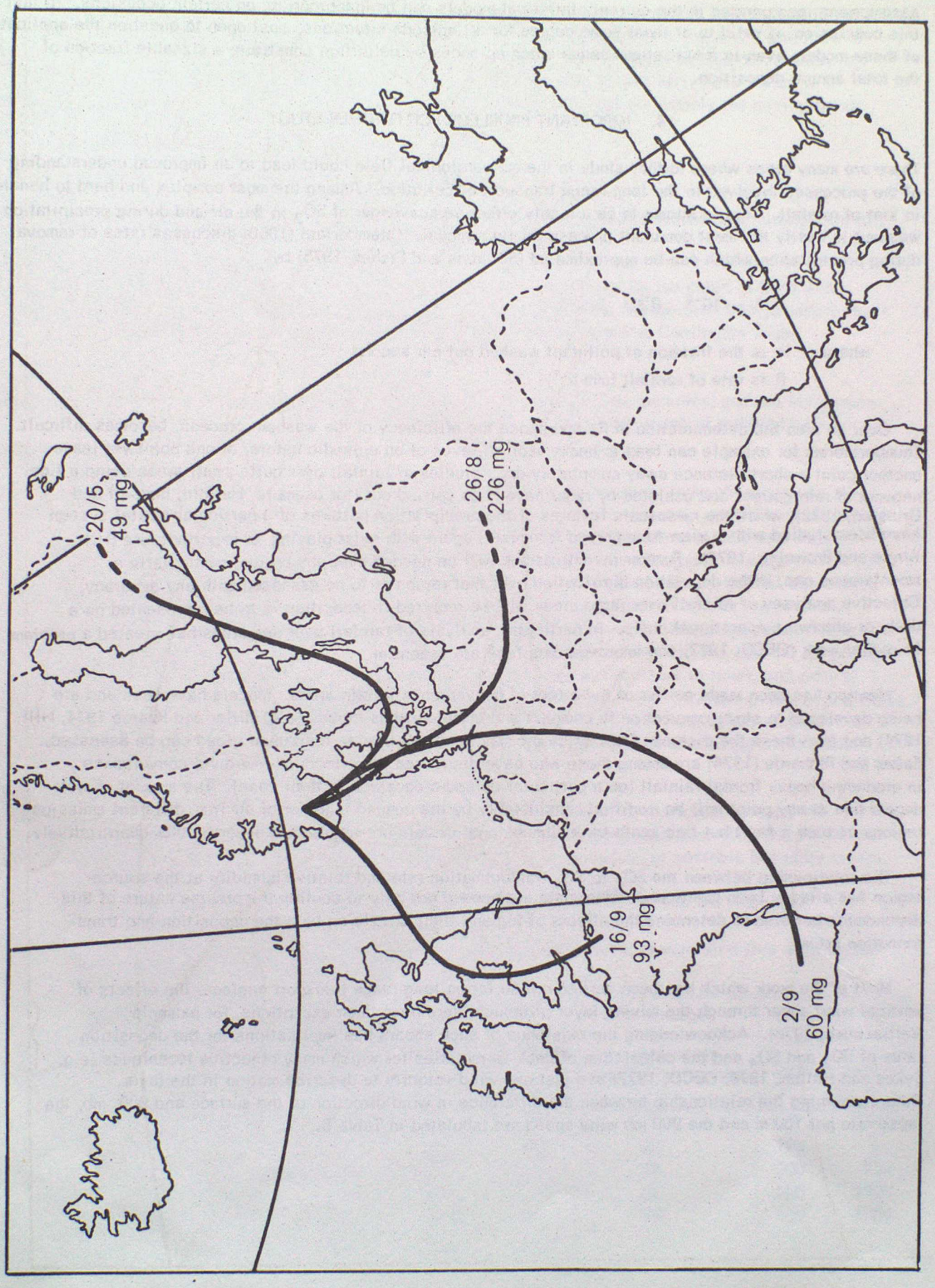

FIGURE 9. Back trajectories for the episode days at Vatnedalen, 1974

assumptions incorporated in the current statistical models can be inappropriate on certain occasions. In fact this conclusion, if valid to at least some degree for all episode situations, must open to question the application of these models even in a statistical sense since episodes by definition constitute a sizeable fraction of the total annual deposition.

4. IMPORTANT PROBLEMS FOR FURTHER STUDY

There are many areas where further study in the meteorological field could lead to an improved understanding of the processes involved in the long-range transport of pollution. Among the most complex and hard to handle is that of rainfall. This is known to be a highly effective scavenger of SO_2 in the air and during precipitation washout is easily the most dominant process for its removal. Chamberlain (1960) discusses rates of removal during precipitation which can be approximated (Scrivens and Fisher, 1975) by

$$\lambda = 10^{-4} \quad R^{\frac{1}{2}}$$

where λ is the fraction of pollutant washed out per second

R is rate of rainfall (mm h^{-1})

Over an area the determination of R, and hence the efficiency of the washout process, becomes difficult. Thunderstorms for example can lead to heavy rainfall, even of an episodic nature, at one point yet leave another point a short distance away completely dry. Studies of rainfall over quite small areas using a fine network of rain gauges and assisted by radar have been carried out (for example, Harrold, Bussell and Grinsted, 1972), while the mesoscale features of the precipitation patterns of a particular frontal system have been studied with a view to improved forecasts, again with radar playing an important role (Hill, Whyte and Browning, 1977). Further investigations will be needed over any region, particularly a mountainous one, if the deposition distribution over that region is to be assessed with any accuracy. Objective analyses of rainfall over large areas will be required if deposition is to be represented on a daily or otherwise operational basis. In particular, analysis of rainfall over sea areas has created a problem in earlier work (OECD, 1977) and improved data here are essential.

Mention has been made earlier of the effect of convergence in rain areas. Models have been and are being developed to study convection in cumulus and cumulonimbus clouds (e.g. Miller and Pearce 1974, Hill, 1974) and from these the distance from which air may be drawn into an individual cloud can be assessed. Salter and Richards (1974) are among those who have discussed the effects of low-level convergence in producing heavy frontal rainfall (on a particular episodic occasion in their case). The amount of wet deposition at any point will be modified considerably by the coming together of air from different emission regions at such a front but fine scale three-dimensional models are required to describe this quantitatively.

The relationship between the SO_2 to SO_4 transformation rate and relative humidity at the source region has already been mentioned. More data are needed not only to confirm the precise nature of this dependance but also to determine the effects of fog and light drizzle on both the deposition and transformation rates.

Most of the work which has been carried out so far on long-range transport neglects the effects of vertical wind shear through the mixing layer (although there have been exceptions, for example Veltischeva, 1976). Acknowledging the existence of such shears has implications for the deposition rates of SO_2 and SO_4 and the calculation of back-trajectories for which many objective techniques (e.g. Sykes and Hatton, 1976; OECD, 1977) use just one wind velocity to describe motion in the layer. Data concerning the relationship between the difference in wind direction of the surface and 900 mb, the lapse rate per 100 m and the 900 mb wind speed are tabulated in Table 6.

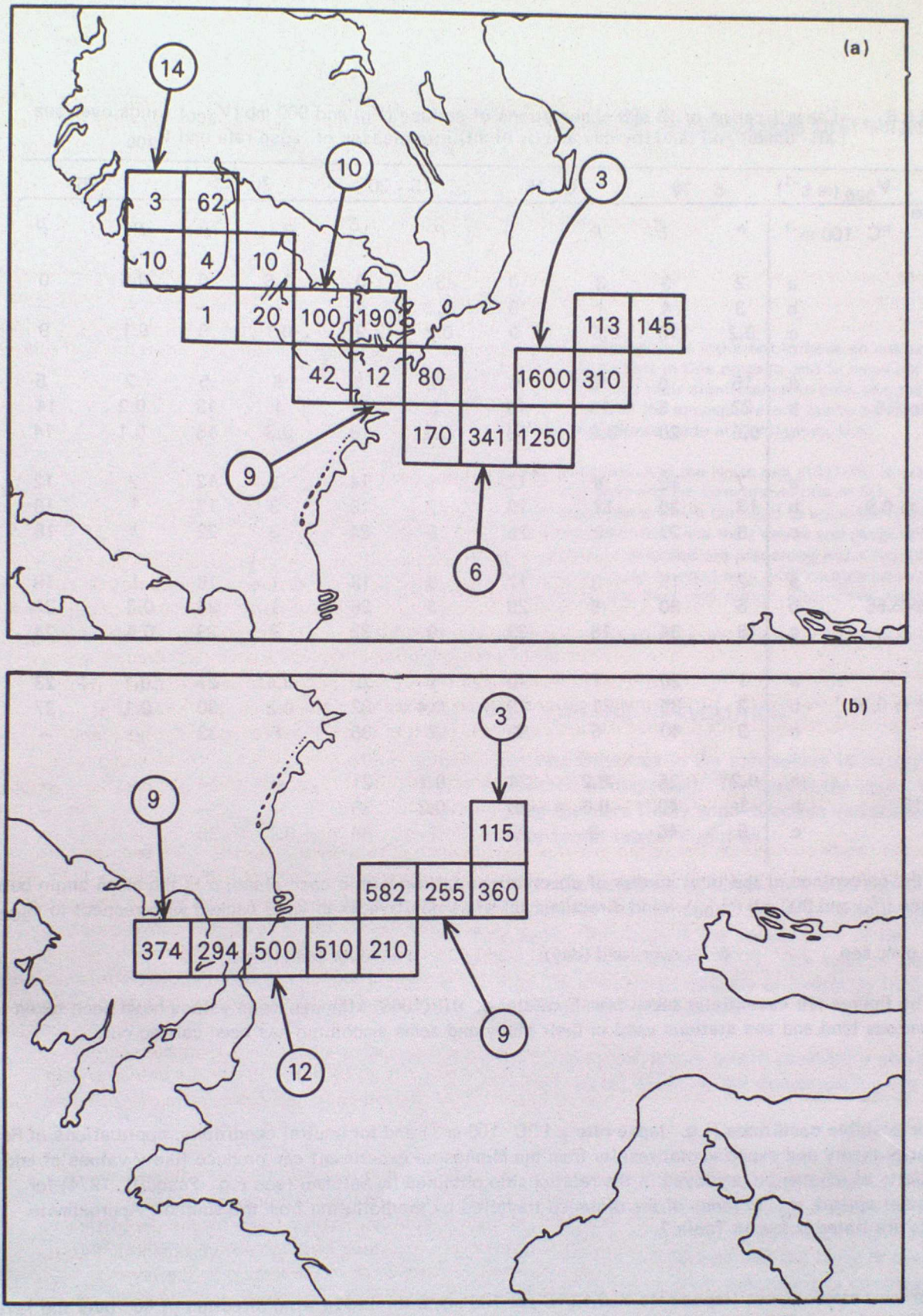

FIGURE 10. Emission squares and emissions (kilotonnes per year) crossed by the back trajectories from

(a) Vatnedalen on 00 GMT, 27 August 1974, and

(b) Cottered on 8 August 1974

Numbers in circles refer to surface geostrophic winds (m s^{-1}) at various points on route.

TABLE 6 Classification of 16 898 observations of surface (V_0) and 900 mb (V_{900}) winds over sea (all times) and land (by day and by night) into classes of lapse rate and V_{900}

	V_{900} (m s^{-1})	5 - 10		10 - 15		15 - 20		20 - 25		> 25	
lapse rate °C 100 m^{-1}		P	$\bar{\theta}$	P	$\bar{\theta}$	P	$\bar{\theta}$	P	$\bar{\theta}$	P	$\bar{\theta}$
	a	2	0	3	0	3	0	1.5	0	0.5	0
≥ 1	b	3	- 4	1	5	0.2	4	–	–	–	–
	c	0.2	10	0.6	9	0.2	9	0.1	9	0.1	9
	a	15	5	16	6	12	6	6	5	2	5
0.9 to 1.0	b	22	5	13	10	5	11	1	13	0.2	14
	c	0.5	20	0.2	19	0.2	18	0.3	16	0.1	14
	a	7	10	8	12	5	14	3	12	2	12
0.55 to 0.9	b	13	20	12	19	7	18	3	17	1	16
	c	6	27	9	26	8	24	3	22	1	18
	a	2	15	3	17	3	18	1	18	1	19
0.2 to 0.55	b	5	30	5	29	3	26	1	24	0.3	21
	c	18	34	18	33	9	32	2	29	0.4	24
	a	1	20	1	20	1	20	0.4	21	0.1	23
-0.12 to 0.2	b	2	35	1	33	0.4	32	0.2	30	0.1	27
	c	9	40	5	39	2	35	1	32	–	–
	a	0.2	25	0.2	24	0.1	21	–	–	–	–
≤ -0.12	b	1	40	0.3	38	0.2	36	–	–	–	–
	c	5	45	3	42	1	38	0.1	35	–	–

P is the percentage of the total number of observations falling within each class, $\bar{\theta}$ is the mean angle between surface (θ_0) and 900 mb (θ_{900}) wind directions taken as positive when θ_0 is backed with respect to θ_{900}.

a = over sea; b = over land (day); c = over land (night)

The figures are essentially taken from Findlater et alii (1966) although mean values have been taken for the various land and sea stations used in their study and some smoothing has been carried out.

For unstable conditions (i.e. lapse rate ≥ 1 °C 100 m^{-1}) and for neutral conditions, applications of Rossby similarity theory and experimental results from the Minnesota experiment can produce likely values of eddy diffusivity which can be employed in the relationship obtained by Saffman (see e.g. Pasquill, 1974) for horizontal spread, σ_y, in terms of the distance travelled by the pollution from the source. Approximate results are listed below in Table 7.

For very stable cases (lapse rate < -0.12 °C per 100 m), a turning in wind direction of 40° over the layer from the surface to 900 mb can be taken as representative. The calculations assume that pollution uniformly fills the relatively deep layer even though the layer is now non-turbulent having been diffused during the previous day-time period when vertical turbulence was relatively vigorous. Since the 'particles' of pollution now remain at virtually a constant level, wind direction shear has a maximum effect. On the Gaussian assumption that σ_y is approximately 1/6th of the total width, the figures in the final column of Table 7 are readily deduced.

TABLE 7 The horizontal spread (σ_y) caused by wind shear alone, related to distance from source (x).

x	σ_y	
	unstable & neutral	stable
km	km	km
1	0.1	0.2
5	0.3	0.8
10	0.5	1.7
50	1.0	8.3
100	1.5	16.7
1000	4.7	167

A mixed layer of 1 km is assumed. The average wind speed is 7.5 m s^{-1} and the stability is assumed to be constant (an implausible assumption, in fact, beyond 100 km).

Clearly, the effect of wind shear on horizontal spread is greater, using these figures and assumptions, in very stable conditions than otherwise. Nevertheless, even in this case, the width of the resulting plume after 100 to 1000 km is similar to that which would be achieved with the amount of synoptic swinging of the back-trajectories normally encountered over several hours. In unstable or neutral conditions, the effect of wind shear is found to be very small. In conditions of penetrative convection, however, pollutants could be taken to heights much greater than 900 mb provided precipitation is not falling and washing out the sulphate. Once any convective cloud evaporates, the remaining sulphur could be strongly affected by the different wind velocities found at these greater heights and would lead to a much greater horizontal spread than the values given above. The fate of pollutants once above the mixing layer has been largely ignored in long-range transport studies, but this may need to be remedied in the future.

Both the path and the spread of pollution released into the atmosphere at or near ground level are also modified drastically by the topography of the underlying surface especially in stable conditions. Numerical techniques such as those described by Mason and Sykes (1977) are currently being developed to study the effect of mountain ranges on boundary layer flow and they could be also valuable in the long-range transport field.

To sum up then, studies of the general precipitation problem, of the effects of wind shear, combined with the diurnal cycle of stability, on lateral dispersion and of the general effects of topography are the most pressing problems which require further study. Meteorological aspects also enter more indirectly into other problems, such as the calculation of deposition and transformation rates both of sulphur compounds and other chemicals, but these are outside the scope of this paper.

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