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the United Kingdom

by

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Airborne Transport of Sulphur Dioxide from the United Kingdom

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Summary

Sulphur dioxide and sulphate have been sampled from aircraft flying along specified tracks off the east coast of England in relatively simple westerly meteorological conditions. The implied fluxes have been related to estimated emissions of sulphur dioxide into the air as it passed earlier over the source regions of Britain. The results imply that typically about 30% is lost by dry deposition to the surface before reaching the tracks, corresponding to a velocity of deposition about 0.8 cm s^{-1}, a further 50-60% typically leaves the country as sulphur dioxide in dry conditions and the remainder as sulphate. The fraction converted to sulphate appears to depend quite sensitively on relative humidity. In rainy situations not only is this fraction quite high but the subsequent wash-out appears to be highly effective.

Introduction

Sulphur dioxide gas is released to the atmosphere in many industrial and domestic processes, mainly as a result of burning fossil fuels, in quantities that are questionably a hazard to the environment. Indeed over a decade ago the issue was raised that as a result of this release by the principal industrial nations of Western Europe, the lakes and rivers of southern Scandinavia were becoming acidic to the point that fish populations were being severely depleted. Although even today the real reasons for this established depletion are not entirely clear it seems that the following play an important role in that area, namely modern forestry practice of planting, drainage and application of fertilizers, the inherently rather acidic nature of the thin layer of soil and, as initially suggested, the fallout (dry deposition and in precipitation) of sulphuric acid, derived from the sulphur dioxide by oxidation, and other strong acids.

What is still not absolutely clear is the extent to which the acidic fallout is responsible for the problem and also what fraction of this fallout comes from nearby sources in Scandinavia and what fraction comes from distant sources, and the United Kingdom in particular (lying upwind in the supposedly predominant S.W. flow). Nevertheless it has become obvious that sulphur dioxide can travel considerable distances in the atmosphere, that it is deposited to the land or sea surface at a rate $v_d \times C$ where C is the low level concentration and where v_d is a velocity of deposition of the order of 1 cm s^{-1}, that it is converted to sulphate as a result of rather complex chemical processes in typical industrially polluted air at a rate which appears to depend rather critically on relative humidity, that sulphate particles can easily be lost to the ground during precipitation, and that in consequence a small but possibly significant fraction of southern Scandinavia's air-derived sulphur burden does indeed come from the United Kingdom. It has also been argued that other areas such as the Ruhr, DDR, Poland and Russia must also make significant contributions of similar magnitudes.

Perhaps if it were only the fish in the lakes and rivers that were being affected, distressing though such ecological damage is, the political pressure to find the cause and a solution would not have been so great. However at one stage it was feared by the Scandinavians that the yield of their economically important forests was also being adversely affected. The evidence for this while still in dispute, is generally considered to be rather tenuous although no-one can deny that a possible risk does exist and that sulphur dioxide emissions should not be allowed to increase greatly without some kind of control and that the effects should be carefully monitored.

Consequently in June 1969, the OECD set into motion a series of expert meetings and planning exercises to assess the problem and to formulate a plan of action acceptable on a cooperative basis by the members of OECD. By the Autumn of 1971 an acceptable plan had been arrived at and was approved by the OECD Environment Committee. After further administrative problems had been solved the OECD Council finally accepted the plan in April 1972 with participation by Austria, Denamrk, Finland, France, Germany, the Netherlands, Norway, Sweden, Switzerland and the United Kingdom, with the overall measurements and studies to be coordinated by the Norwegian Institute for Air Research (NILU) situated at Kjeller, near Oslo.

The United Kingdom felt it could best contribute to the solution of the problem by additionally developing its own independent programme in which certain key questions have been selected, studying these in detail and where appropriate mounting actual experiments specifically designed to provide the required answers. One of the major questions was concerned with the rate at which sulphur dioxide emitted into the atmosphere was lost by dry deposition to the ground over typical countryside or to the sea surface, or was converted to sulphate by chemical reactions within the atmosphere. These are the questions considered in this paper, being clearly relevant to the specific question as to how much sulphur released

by the major industrial sources of Europe reaches Scandinavia and is deposited there. The problem could be subdivided into five key parts:

(a) derive a suitable small-scale emission map of sulphur dioxide from all sources for the UK.

(b) mount suitable sulphur dioxide and sulphate measuring equipment on aircraft capable of flying for sufficient periods of time within the downwind plume at enough heights to enable an accurate measure of the fluxes of both forms of the pollutant to be estimated.

(c) determine areas where flying could be authorized, avoiding busy civil aircraft lanes, military flying or danger zones, or mountainous regions, and within range of useable airfields.

(d) select meteorological situations which will carry the sulphur from the major known source regions to the permitted aircraft-sampling tracks and which are sufficiently simple in character that the results of the measurements can be interpreted with a minimum of uncertainty being introduced by the meteorology.

(e) develop a scheme for deriving sufficiently accurate back trajectories that the fluxes can be directly related to emissions into the same air mass.

The following sections report on these aspects of the problem and on the conclusions that have been reached. The flying was carried out by members of the Meteorological Research Flight at RAE Farnborough and the development of the sampling technique and the actual analysis of the samples by AERE Harwell.

Some similar flights have been made by Warren Spring Laboratory but are not reported here.

Meteorological Conditions

Sampling flights were made in 1971 and, with some changes in plan, again in 1973. The following criteria were applied in forecasting suitable conditions although in the event were on occasions either deliberately relaxed or were simply not attained in practice.

(i) The surface geostrophic wind speed G should exceed 5 ms^{-1} (in 1971) or 10 ms^{-1} (in 1973). The increase in 1973 was made to simplify the calculation of back trajectories.

(ii) The surface geostrophic wind direction θ should lie between 200° and 290° (1971) and be relatively steady for 12 hours over most of England preceding the time of sampling. During 1973 the plans were enlarged to allow for the possibility of sampling the same air twice, once off the East Coast late on one day and then a second time the following morning off the Danish Norwegian coast. For this to be possible θ had to lie between 220° and 260°.

(iii) The turning of the wind should not exceed 10° within the mixing layer.

(iv) The criterion regarding rain until the final flight in 1973 was that there should be no widespread and continuous rain over England and Wales during the preceding 24 hours before the time of sampling. However as confidence grew in the interpretation of the results, this restriction was relaxed and indeed the last flight was made with widespread showers present.

(v) Since sampling could not be carried out effectively in cloud due to potential damage to the filter paper used in the sampling apparatus the area should either be cloud free or have cloud only above the mixing layer. Sampling in rain could be dangerous for the same reason but it was felt that in the one flight with rain no significant losses were in fact incurred.

(vi) The mixing layer should be less than 3 km in depth.

The Sampling Plan

Due to aircraft safety rules, no sampling could take place during hours of darkness. Consequently in 1971 most sampling times were around midday and early afternoon. In 1973 sampling times off the East Coast were delayed until late afternoon or early evening for two reasons. Firstly in order to sample during the high and relatively steady emission values which

at source occur during the 09 Z to 20 Z period, and secondly to make it possible in favourable circumstances to sample in the same air mass off the Danish-Norwegian coast the following morning.

The aim was to achieve an adequate as possible flux profile through the mixing layer. In 1971, and off the Danish coast in 1973, samples were taken at three levels within the layer, one at 150m, another at 150m below the top and a third midway in between. Off the East Coast in 1973, five samples were taken, the extra two being evenly spaced between the other three defined as before. In addition one sample was taken about 300 m above the mixing layer and on the long flight across the North Sea, this was made just prior to arrival at the prescribed sampling track.

At least 500 litres of air had to be sampled to determine the sulphur dioxide content at any particular level, and since this took about forty minutes, the sampling track length had to be about 160 to 180 km for the Varsity aircraft used.

A series of tracks off the E Coast of England, 180 km long and normal to wind directions of WNW, W, WSW, SW and SSW were agreed with the Meteorological Research Flight for the 1971 experiments which avoided civil air lanes and danger zones and which were downwind of the major industrial source areas. The three actually used are shown in Figure 1.

In 1973 the tracks were altered in order to minimise the effect on the estimate of emissions into a back trajectory resulting from possible forecast errors of $\pm$ 20° in wind direction. This was done by considering a sequential series of 40 km long tracks, F to Q, as shown in Figure 3 and, for each wind direction between 210° and 300°, the total integrated sulphur dioxide source strength in thousands of tons per year into the appropriate back trajectories were evaluated assuming a constant uniform wind and using the emission values given in Figure 5. Figure 4 shows how the plumes from major source regions affect different tracks for different wind directions, sometimes overlapping other major plumes to give "local" peaks. Four such tracks each 40 km long had to be combined to give a full sampling track of 160 km. By considering the relative variations in total source strength as wind direction changed by $\pm$ 20°, 160 km-tracks showing minimum response could be

selected for each basic wind direction. The shaded areas on Figure 4 are those for which percentage errors, are less than 25% for $\pm$ 20° wind direction uncertainty, are shown, and the final choice of sampling tacks is shown in Table 1.

The determination of the sulphur dioxide emission distribution

The determination of the distribution and magnitude of sources is a very important step towards a meaningful interpretation of the results of the sampling flights. Fortunately a considerable amount of basic data proved to be available from which fairly reasonable source estimates could be made. At the time of analysis the most up-to-date set of complete data was for 1970 which, although applying to a year prior to the years when the sampling flights were made, could be considered very adequate on the grounds that overall emission trends from year to year have been quite gradual in the last five years.

The main sources of information have been:

(i) The "Digest of Energy Statistics 1971", produced by the Department of Trade and Industry in which are tabulated in some detail the consumption of primary fuels by all the major consumers, industrial, domestic etc and broken down into what are called the twelve Standard Statistical Regions of the United Kingdom. These regions contain several counties or parts of counties; for example "West Midlands" includes Hereford and Worcestershire, Shropshire, Staffordshire and Warwickshire.

(ii) The "Atlas of Britain and Northern Ireland" published by the Oxford University Press and the "Desk Atlas Maps" (particularly No 107) prepared by the Ministry of Housing and Local Government, 1968, contain valuable information on the geographical distribution of the relevant major industries, power stations and oil refineries. This information enabled us to subdivide the emissions from each type of source within each Statistical Region down to individual counties and ultimately to individual 20 km by 20 km squares. Where smaller industries and

domestic emissions were concerned these were divided up according to population within the region, thereby assuming some kind of uniformity of fuel use.

(iii) "The National Survey of Air Pollution, Report No 1", May 1970, prepared by Warren Spring Laboratory (Department of Trade and Industry) and a draft copy kindly supplied by the author M L Weatherby, of "Fuel consumption, and smoke and sulphur dioxide emissions and concentrations in the United Kingdom" (May 1974) to appear in the same Warren Spring Laboratory series. These papers enabled the fuel consumptions by the various consumers to be converted to emissions of sulphur dioxide, taking into account both the sulphur content of the fuels used and also the retention, where appropriate, of sulphur in the ash or, in the case of iron foundries, in the iron itself.

Most of the essential information is included in Tables (2) and (3) and in Figure (5). These figures have been confirmed as broadly correct, wherever a check is possible, by representatives of some of the other appropriate Departments, for example by Warren Spring Laboratory, although no doubt there may remain some inconsistencies which are hopefully only minor in nature.

Table (2) shows the basic facts for coal, solid smokeless fuels and petroleum products subdivided on a broad consumer basis and gives the total consumption of each and what this means in terms of final sulphur dioxide emission to the atmosphere. The largest single contributor to the atmosphere's load comes from power station emissions, roughly 2 million tons per year from coal-fired stations and over half a million tons from oil-fired ones. The sum of these is probably more constant in these days of uncertain fuel supplies than the individual components. The next largest contributor is industry with about 2 million tons SO_2 per year. The total emission for industry, domestic sources etc is around 5.8 million tons or, on a very rough basis, an average of 2 cwt or 100 kg SO_2 per member of the population. This is both an interesting and useful figure since as one of us has shown

(Smith, 1973) if we exclude the contribution from the known and very localised power stations the emission from any town can be estimated quite accurately over a year or more on the basis of 50 kg SO_2 per person times an easily assessed "Industrial factor" which varies from 0.5 in non-industrial residential areas, or country towns, to 2.0 in towns with heavily-polluting industy.

Table 3 summarises the break-down of emissions on a county and consumer basis. The industrial break-down has been abbreviated in the Table, for reasons of space, to include only "iron and steel manufacture", "engineering" and "the chemistry industry", leaving all other sources grouped together in "other industry.". In Norther Ireland, since the corresponding values were not so readily available and since most of the heavy industry appears concentrated within the Belfast area, no breakdown by industry is given.

The location and capacity of the power stations and oil refineries are known from the Tables and Maps quoted above and the consumption of fuel and corresponding emissions of SO_2 have been deduced on the assumption of proportionality to their respective capacities. All values in the Table are in thousands of tons SO_2 per year released to the atmosphere.

For our purposes some of the counties are too big to enable reasonable estimates of emissions into the back trajectories to be easily made. Consequently a further break-down was made into 20 km by 20 km squares. The results are shown in Figure (5). The squares correspond to the standard Ordnance Survey grid squares. Put into this form an immediate visual impression is gained of the most important source areas, which of course correspond directly with the major industrial areas of the country but also show where river-water supplies are sufficiently adequate to support major power station installations eg along the Nottinghamshire Rivers Trent and Soar. The largest source regions are shown to be London and the Thames estuary, Merseyside and South Lancashire, Southwest Yorkshire and Nottinghamshire, Glamorgan and Monmouthshire coastal belt, Newcastle and Durham, and Lanarkshire.

The back trajectories

In the analysis of the results one of the hardest problems, and one of the most important, is to obtain reasonably accurate back trajectories so that the measured fluxes may be related to the real emissions upwind. Errors can occur in both speed and direction of the advective wind used to get these trajectories. These errors are inter-related especially in a changing synoptic pattern. However in a direct sense, errors in speed only give apparent emissions that are unrepresentative of the true time of passage over the sources and which arise from the usual diurnal variation in emission rates. Errors in direction can be more serious, distorting the trajectory paths over source regions that do not in reality affect the concentrations along the sampling tracks. As described in the section on selected sampling tracks an attempt was made to minimize the effect of direction errors by choosing tracks for a forecast wind direction on "plateaux" of the total upwind source strength versus wind direction map (Figure 4) where small errors in the forecast direction and in the estimated back trajectory will not radically change the total input emissions.

The major problem is of course that the wind in the boundary layer often varies in a way that cannot be explained by simple horizontally-homogeneous stationary boundary layer theory. These variations may arise from temporal changes in the synoptic pattern, from variations in the underlying topography and from diurnal variations in stability in the lower layers.

Usually the only time winds measured in the airmass being sampled are those made by the aircraft. Our approach has been to relate these in some meaningful way to the surface geostrophic winds and to the surface 10-metre winds when these have been available. Several plausible relationships were tried out on the 1971 data and generally the acrosswind separation between tracks over significant source areas in Britain did not exceed 30 miles and was often considerably less in these relatively simple meteorological situations. Subsequently the following relatively simple procedure was used; a mean wind normal to the track was formed from the measured aircraft 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 was compared with the local surface geostrophic wind speed G, and the ratio 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-metre wind direction θ_s and the ratio $(\theta_G - \theta_p)/(\theta_G - \theta_s)$ was also assumed constant along the back trajectory.

Tables (4) and (5) present the basic data for the seven flights. In retrospect one interesting consistency is apparent; the ratio V/G and the angular difference $(\theta_G - \theta_p)$ are both well related to the overall stability of the mixing layer, for when this is near neutral or slightly unstable

$$V/G \approx 1 \quad \text{and} \quad (\theta_G - \theta_p) \approx 10^\circ$$

whereas when the layer is slightly stable

$$V/G \approx 0.9 \quad \text{and} \quad \theta_G - \theta_p \approx 20^\circ$$

The estimated mean trajectories using the constant ratio method are displayed for the seven flights in Figures 1 and 2, together with the hours of passage and a brief description of the current weather. The winds are appropriate surface geostrophic winds over land and differ a little from those estimated along the sampling tracks. An estimate of the emissions into the sampled air can be deduced fairly simply by considering the contribution of each source within the trajectory limits. On the basis of a constant ratio V/G and neglecting any net vertical velocity of the air, each source contributes an equal time's worth of sulphur dioxide, the emission over one hour, to the airmass that takes one hour to pass the sampling track, and this is true regardless of whether the speed of the airmass quickens or slows along its trajectory across the country. This property makes the calculation relatively simple; the emission value associated with each 20 km by 20 km square lying within the bordering trajectories (see Figure 5) is included and has only to be weighted by two factors, the diurnal and date factors which are derived from data on electricity consumption but are assumed

to hold for all types of sources. The diurnal factor depends solely on time of day (see Figure 6) reaching a peak at 10 Z and remaining high until late evening when it falls rapidly to the early-hours' trough at 04Z. This factor will therefore vary along the trajectory unlike the date factor which is constant for any one flight. The date factor curve demonstrates the typical source variation with time of year, but since emissions in general and domestic heating sources in particular will vary with temperature, the weighting factor is taken for that day whose climatic average temperature is the same as the actual average temperature on the day of the flight.

In 1973 a refinement was used that was absent in the 1971 flight analyses. Since sampling at various heights along the track is spread over 2 or more hours the wind may be changing and the appropriate back trajectories and corresponding emissions may be somewhat different for the low and the high sampling levels. To allow for this, separate back trajectories were obtained for each level and the appropriate flux of sulphur dioxide was then divided by the emission into the back trajectory determined for that time. These ratios were plotted and an integrated flux to emission value obtained for the whole sample flight. These values appear in Table (6).

Sampling and chemical analysis

The Meteorological Research Flight of the Meteorological Office in collaboration with the Health Physics and Medical Division of the Atomic Energy Establishment, Harwell, has developed a filtration system for the simultaneous determination of sulphate aerosol and sulphur dioxide concentrations from an aircraft using a discrete method with post-flight chemical analysis of the samples. The technique involves passing the air containing sulphur dioxide through a filter paper impregnated with a chemical absorbent and subsequently analysed as sulphate. Sulphate aerosol which would otherwise interfere with this has to be first removed with high efficiency filters. This is done by using two filters, a cellulose Whatmen 41 filter placed in front of a "Microsorban" absolute pre-filter, both of which have acceptably low uptake of sulphur dioxide under sampling conditions. The sulphate-free

air then passes through a further filter paper of low sulphate background (Whatman 41) impregnated with a solution of 25% weight by volume Analytical Reagent grade potassium carbonate in 10% glycerol solution. In preparation the paper after impregnation was squeezed through rubber rollers to remove excess reagent and then quickly dried by infra-red lamps. This filter provides a very adequate absorber of the sulphur dioxide and gives reasonable accuracy above a concentration of 3 $\mu g m^{-3}$.

The filters and their holders were installed on two booms extending from the Varsity aircraft through openings in the roof and port side, ahead of the engines. Either boom could be connected to a reciprocating diaphragm air pump by means of a system of valves and pipes incorporating a special orifice flow meter. Normally each filter was exposed for 10 minutes, representing about 40 km distance flown, during which time 0.5 m^3 of air was sampled. During flight a filter pack was secured in the sampling head and the boom extended. The booms were used alternatively in this way to obtain successive samples on a long flight path. When sampling in cloud the central area of the paper prefilter would become wet and occasionally punctured, but the "Microsorban" filter remained intact. Possible losses of sulphur dioxide may have occurred under these conditions due to absorption on the wet filter material.

Before a flight several packs were loaded under clean conditions and stored in aluminium containers or polythene bags. Certain of these, called control filters, were not exposed and were subsequently analysed for each flight as a check. A further batch of 8 loaded but unexposed filter packs stored in such bags for 55 days gave a mean level 1.2 μg SC_2 (max 1.4) on the paper.

After the flights the sulphur dioxide was retained as potassium sulphite which was gradually oxidised to sulphate, the process being completed and the sulphate removed by hydrogen peroxide solution. Following Persson's thoronal method the solution was passed through a column of cation exchange resin which removed the potassium ions. The solution would then rapidly settle to a suitable pH for analysis without further adjustment.

Particulate sulphates collected on the cellulose prefilters could be extracted with water and determined by the thoronal method without further treatment.

Further details are given by Healy and Atkins (1974).

Interpretation of the results

The results of the seven sampling flights off the east coast of England are set out in Table (6). The first part of the Table gives some of the basic properties of the sampled 'plume' and relevant meteorological factors; h is the estimated depth of the mixing layer deduced from nearby radio-sonde ascents taken at the main synoptic recording stations, Hemsby, Shanwell, Stornoway, Crawley, and Aughton, and modified as necessary by observations of haze-top made from the varsity aircraft just before sampling and of temperature. The plume of sulphur dioxide in most cases had not completely filled the mixing layer, particularly on the somewhat stable days (see Table 4) and this is confirmed by the magnitude of σ_z (the root mean square value of height of the concentration distribution) which in many cases is significantly less than $h/\sqrt{3}$, the value for a uniform distribution .

$$\sigma_z^2 = \frac{\int_0^\infty z^2 C(z)\, dz}{\int_0^\infty C(z)\, dz}$$

where C(z) is the sulphur dioxide concentration at height z. Only on October 22nd, 1971, was the fairly shallow mixing layer virtually filled out at this range from the sources.

V is the mean speed of the pollution over the distance x from the upwind major source to the sampling track and agrees fairly well with the mean plume speed at the sampling line given in Table (5).

The expected neutral value of σ_z is also given. This is the value assuming neutral stability and travel over typical British countryside using the modified form of Pasquill's vertical diffusion scheme given by Smith (1972). Provided vertical spread has not been limited by the top of the mixing layer, the ratio $\sigma_z / \sigma_{z\,(neut.)}$ is an indication of the overall stability of the layer, values below 1.0 would suggest a stable situation,

values above an unstable situation. Comparison with the subjective assessments of the stability from the vertical temperature profiles given in Table (4) generally confirm this although the supposedly unstable situation on Oct 8th 1973 has too small a value of σ_z .

The last entry in the first part shows whether or not any significant rain had fallen. Only on the last flight was there any, when, although only occasional showers had been noted over land, by the time sampling started rain was fairly widely affecting the airmass; on the lower two tracks between 1350 Z and 1510 Z slight rain was falling from medium cloud and, by 1515 Z when the third leg commenced at 828 metres moderate rain was falling over the middle third of the track.

The next part, a single row, gives the estimated emissions E_2 of sulphur dioxide into the sampled airmasses using the back trajectories shown in Figures (1 & 2), the source map (Figure (5)) and the date and time weighting functions given in Figure (6).

The third part of the Table provides the data concerned with the sulphur dioxide, measured or inferred. F_2 is the measured flux of SO_2 across the track using the data in Table 5 measured in thousands of metric tons per hour, assuming background values are negligible . The ratios F_2/E_2 give the fraction of emitted sulphur dioxide remaining in the atmosphere as SO_2 along the track. The fractions vary from 0.34 on 8th October 1973 to 0.71 on 22nd November 1971 and do not obviously appear to be correlated with distance x or time of travel t. However this is somewhat of an illusion since on the first three flights of 1973 when the sulphate flux F_4 was also measured and no washout had occurred, simple budgetary considerations could yield the fractional amount of SO_2 that must have been lost by dry deposition before the sampling track was reached. This is given by the amount D_2/E_2 where D_2 is the total dry deposition of SO_2. The budget requires $F_2 + F_4 + D_2 \equiv E_2$. This loss can be assessed using a simple developing plume model in which the plume is supposed to grow in depth with distance at a rate equal to that in neutral conditions, given in Smith's (1972) curves, modified by the known ratio

σ_z / σ_z (neutral) on the sampling track, and assuming that the loss to the ground is proportional to the concentration within the plume, then

$$\frac{dQ}{dx} = - \frac{Q}{V} \frac{v_d}{\sigma_z(x)}$$

where V is the mean wind speed and Q(x) is the flux of SO_2. The equation relates the constant of proportionality, the so-called velocity of deposition v_d, to the total loss by deposition D_2. Often v_d is defined otherwise in terms of the concentration at a specified level close to the surface but this is not possible in our experiments since the first level is normally as high as 150 metres and the tendency for the concentration to decrease below this as a direct result of deposition cannot be easily assessed. However provided the concentration profile does not greatly differ from a Gaussian distribution the two definitions are not likely to produce systematic differences exceeding 10 or 20% with our equation tending to possibly over-estimate v_d. A further complication arises from the conversion of some of the SO_2 to sulphate, the absorption of which at the surface is assumed negligible. As argued below most of the sulphate is produced within the first 50 km from the source. This partially invalidates the above equation and may lead to slight underestimation by some 10-20%. Hopefully the two errors may tend to cancel but a possible uncertainty in v_d of around 10% must remain. Table 7 gives the result of integrating this equation assuming certain specified values of the initial σ_z to allow for the rapid mixing within the building complex and the various chimney emission heights in the source area. For example on the 4th September 1973 from Table 6 $\sigma_z \approx \sigma_z$(neutral) $\approx$ 750 m, x = 125 km, V = 8.1 ms^{-1}, D_2/E_2 = 0.33. Thus 67% equivalent-SO_2 remains and from Table 7 for the above value of x, v_d must range from 1 x 10^{-3} for small $\sigma_z(o)$ to 1.1 x 10^{-3} for larger $\sigma_z(o)$. Thus v_d = 0.8 cm s^{-1} for small $\sigma_z(o)$ or 0.9 cms^{-1} for larger $\sigma_z(o)$.

On the remaining two "dry" 1973 flights very similar values of v_d are found, ie 0.8 cms^{-1} and 0.6 cms^{-1}. The last situation was off the Durham coast when the pollution spent relatively longer over the sea than over land, and the slightly smaller value of v_d may reflect the slower uptake of SO_2,

that has been generally forecast, by the dynamically-smoother sea surface.

In so far as we can feel confident about the mutual consistency of these relatively few values of v_d, similar values may be assumed in the other flights to see if further self-consistencies can be found. Consequently v_d for all the 1971 flights has been taken as 0.8 cms^{-1}, and the value for the last flight which has a similar trajectory to 7th Sept 1973 has been lowered to 0.7 cms^{-1}. Obviously there must be an uncertainty here of at least $\pm$ 0.2 cms^{-1}. With these assumed values of v_d Table (7) may be used in reverse to complete the lists of values of D_2/E_2. It is probably fortuitous that all these ratios, except one, lie in the narrow range 0.31-0.33.

On the 22nd November 1971 the mixing layer was quite shallow and the evidence suggests that the plume on this occasion had filled the mixing layer for quite a high proportion of its travel from the major source region. Consequently in the determination of D_2 the above developing plume model was replaced by a simple "box" model in which the SO_2 is assumed well mixed through the whole layer. The final row in this part of Table (10) gives the observed average values of sulphur dioxide concentration within the plume along the sampling tracks. These values are relatively low compared to some measurements exceeding 100 $\mu g\ m^{-3}$ measured over the North Sea in slow-moving anticyclonic easterlies bringing SO_2 from the industrial areas of central Europe.

The next part presents data on the sulphate; all values have been expressed here in terms of the equivalent sulphur dioxide to simplify budgetary consideration. Thus 1 ton SO_4 is equivalent to (and is presented in the table as) $\frac{2}{3}$ ton SO_2. The flux of sulphate F_4, corrected for background values obtained from concentrations measured above the mixing layer, was measured in the 1973 flights, and in the case of the first three of these, have already been used above to deduce D_2 and v_d. In the fourth 1973 flight, the same budget could not be applied because some of the sulphate generated had almost certainly been washed out by the rain. If this amount is called F_4 then the new balance must be

$$F_2 + D_2 + F_4 + R_4 = E_2$$

The only unknown on the 8th of October is R_4 and hence can be deduced. It appears that, if the assumed value of v_d is correct, roughly 75% of the sulphate produced was removed by the rain showing just how effective it can be as a cleansing mechanism. The implied total production P_4 on this occasion is given as 34% of the SO_2 emission E_2.

An important problem is to understand if possible how much sulphate will be produced on any one occasion. The chemistry of the process is dauntingly complex and our measurements are totally inadequate to reveal or simulate the transformation processes. However one point immediately became apparent: the amount of sulphate produced seemed, at these values of x, to be poorly correlated with either x itself or t, the time of travel. This seems to imply that possibly most of the conversion takes place very early on near the source region where the concentrations of both SO_2 and the other industrially-produced pollutants that may be involved in the transformation processes will be high. The only component involved in the process that is therefore likely to vary in an important way in these industrial environments from occasion to occasion is the relative humidity and hence a relationship between this and the relative production P_4/E_2 was sought. Figure (7) shows the seven data points with the vertical bars denoting the possible spread in relative humidities along the trajectory implied from measurements made by the aircraft and from relevant radiosondes made upwind over land. Whether it is simply fortuitous or not there does seem to be a definite trend and the curve is an attempt to provide an optimum fit by eye. The conversion rate is seen to increase with relative humidity from a very small value, perhaps zero, at a relative humidity near 60%. P_4^*/E_2 in the Table gives the empirically implied relative production rate using the curve and the 7 values differ rather insignificantly from the rates implied from direct measurements or by budgetary implications. This second consistency over the 1971 date tends to confirm the earlier assumption of assuming $v_d = 0.8$ cm s^{-1}.

The final two rows simply present the basic budget statements, the first is an identity since it has been used to derive D_2 or P_4, whereas the second shows how closely a complete budget balance has been achieved using the empirical P_4* production rates.

Flights over the Danish-Norwegian coast tracks

In 1973 flights were proposed on the far side of the North Sea, along the track line shown in Figure (3), in order to assess the further depletion of the sulphur content of the atmosphere after crossing roughly 200 km of sea. Ideally it was hoped to sample the same airmass both off the E Coast and over the far side. However as one might fear this ideal was never realised. Wind direction and strength forecasts were rarely accurate to the required degree of precision and in fact the only second flight that was thought worth making was on the 7th to the 8th of September as shown in Figure (3). Even here the two sampled airmasses were significantly different. Over this greater range and particularly over a data-sparse area like the North Sea evaluation of back trajectories is somewhat more uncertain, but all the indications suggest that the track went well to the north of that sampled off the Durham coast the previous evening. Interpretation is therefore quite difficult. The basic data are as follows:

Estimated SO_2 flux = negligible

Estimated sulphate flux: 19.9 th^{-1}. Typical conc. in mixing layer 2.5 $\mu g/m^3$ and 0.5 $\mu g/m^3$ above.

Sulphate flux in terms of equivalent SO_2: 13.3 th^{-1}

Estimated emission from track as indicated on Figure (3) = 12.4 th^{-1}

Relative humidity along track: within range 76-92% (average 84%)

Implied conversion of SO_2 to sulphate: 28% (see the last section and Figure 7)

Expected loss of SO_2 by deposition to surface: approximately 50% (also see the last section)

The difficult aspect of these results is that virtually no sulphur dixoide was measured. Three possibilities exist; the first is that the relative

humidity at some stage rose to nearly 100% when almost all the SO_2 was converted to sulphate and then fell again. This would explain the approximate balance between the sulphate flux and the emission of SO_2. The second possibility is that roughly half the pollution from the lowlands industrial area diffused laterally into the sampled airmass. This would account almost exactly for the measured sulphate flux (taking the implied conversion rate) but not for the observed absence of SO_2. The third possibility and perhaps the most likely is that the back trajectory went even further north than indicated, had very small emissions into it, explaining the low SO_2 values, and that the sulphate flux came from natural input from the sea surface. Unfortunately no measurements were made of the other sea-derived ions such as sodium and magnesium to confirm this, but the concentrations were of the same general magnitude as found in the background values above the mixing layer in some of the other flights, these values presumably having come from Atlantic Ocean natural emissions.

Conclusions

The aircraft flights sampling sulphur dioxide and sulphate in relatively simple meteorological conditions off the east coast of England have indicated that meaningful conclusions can be reached regarding the long-range travel of these pollutants. In particular the loss of SO_2 by dry deposition takes place at a rate given by the velocity of deposition v_d which probably lies between 0.8 and 1 cm/sec over land and less certainly between 0.8 and 0.6 cm/sec over the sea. Some fraction of the SO_2 will be converted to sulphate in the relatively polluted environment of the main industrial sources which depends primarily on the ambient relative humidity of the atmosphere. Any later and more gradual conversion has not shown up in our experiments. Rain has been shown to be a most effective scavenger of sulphate on one of the flights removing 75% of that generated in a relatively short distance.

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TABLE 1 Sampling tracks for specified wind directions and minumum error for ±20° wind direction uncertainty.

Wind direction	E. Coast Track	Source Area	Danish-Norwegian Coast Track
220°	IM	Sheffield-Midlands	AC
230°	JN	Sheffield-Notts	BR
240°	KO	Lancs-Midlands	RT
250°	FJ*	Durham	BR
	MP+	Midlands	-
260°	GK*	Durham	RT
	MP+	Midlands	-
270°	HL	Durham	-
280°	IM	Durham	-
290°	JN	Durham	-
300°	KO	Durham-Glasgow	-

+Best track for E. Coast flight only.

*Best track for E. Coast and Danish-Norwegian coast pair of flights.

TABLE 2

1970 Figures for Sulphur Dioxide Emissions to the atmosphere within the United Kingdom

FUEL	CONSUMER	TOTAL CONSUMPTION (million tons)	Reference* (TABLE No)	Assumed** % SO_2 released to atmosphere	Reference*** (Note number to Table 3)	TOTAL SO_2 (million tons)	Emission counted in I: Industrial D: Domestic P: Power Stations O: Oil Refineries
COAL	INDUSTRIAL	23.0	29	2.52	B2	0.58	I
	DOMESTIC (incl schools, hospitals, railways etc)	19.9	29	2.08	B1	0.41	D
	POWER STATIONS	76.0	29	2.7	B3	2.05	P
	COKE WORKS	24.9	29	0.4	A	0.10	I
	GAS WORKS (incl patent fuel & carbonisation plants)	8.3	29	0.72	B4	0.06	I
SOLID SMOKELESS FUELS	DOMESTIC	7.3	90	1.60	B6	0.12	D
	INDUSTRIAL: Iron	11.7	86	absorbed by iron		0	I
	Others	1.4	86	2.34	A	0.03	I
	Public Services	1.2	86	2.34	A	0.03	D
	Miscellaneous	1.5	86	2.34	A	0.03	D
PETROLEUM	FUEL OIL (i) Power Stations	12.4	45,69,41	5.2	A & B	0.64	P
	(ii) Oil Refineries	0.5	41	5.2	A & B	0.02	O
	(iii) Other Industrial	25.0	41	5.0	A & B	1.25	I
	MOTOR SPIRIT	14.0	41	0.1	B8	0.01	D
	DIESEL FUEL	5.0	41	0.6	B9	0.03	D
	GAS OIL	11.3	41	1.4	B10	0.16	D
	(Oil-) REFINERY FUEL	5.9	40,45	3.9	B13	0.23	O
	KEROSINE BURNING OIL (excl aircraft fuel)	2.4	41	0.77	-	0.02	D
TOTAL SO_2 Emission (million tons)						5.77	

*Table number in "Digest of Energy Statistics 1971" Department of Trade and Industry. Publ by HMSO.

**Allows for partial retention of SO_2 in ash (or at source).

***"A" refers to "The National Survey of Air Pollution, Report No 1", May 1970, Department of Trade and Industry (Warren Spring Laboratory). No note numbers are given.

"B" refers to a draft copy of the paper "Fuel consumption, and smoke and sulphur dioxide emissions and concentrations in the United Kingdom" by M-L. P. M. Weatherley (in the same series), May 1974.

TABLE 3

SULPHUR DIOXIDE OUTPUT 1970

Thousand tons

County	IRON & STEEL MANUFACTURE	ENGINEERING	CHEMICAL INDUSTRY	TOTAL INDUSTRY	OTHER INDUSTRY	Domestic	Power Stations	Oil Refineries	Total
Cumberland	17.1	0.6	3.2	23.9	3.0	9.5	13.4	0	46.8
Durham	56.9	2.5	58.4	136.3	18.5	45.2	22.2	23.2	226.9
Northumberland	39.8	2.5	3.2	52.7	7.2	25.2	87.3	0	165.2
Westmorland	0	0.6	0	1.7	1.1	2.1	1.9	0	5.7
Yorks (N Riding)	0	0	0	1.0	1.0	23.1	14.3	11.6	50.0
Yorks (W Riding)	89.2	6.3	24.1	179.8	60.2	115.4	310.6	0	605.8
Yorks (E Riding)	5.2	0.4	1.3	13.0	6.1	16.1	14.3	0	43.4
Lincs (Lindsey)	10.5	1.7	1.3	19.0	5.5	14.6	16.5	23.2	73.3
Derbyshire	17.6	1.8	-	28.7	15.3	23.8	131.9	0	184.4
Leicestershire	5.8	1.8	4.6	21.3	9.1	20.1	5.8	0	47.2
Lincolnshire	0	1.8	-	2.3	0.5	9.2	5.8	0	17.3
Northamptonshire	2.9	1.8	-	7.5	2.8	11.9	7.6	0	27.0
Nottinghamshire	8.7	1.8	-	24.0	13.5	25.7	297.0	0	346.7
Rutland	0	0	-	1.2	1.2	0.9	0	0	2.1
Cheshire	2.9	2.7	19.8	40.7	15.3	44.1	40.2	24.2	149.2
Lancashire	26.4	24.3	79.3	240.8	110.8	147.7	181.6	11.6	581.7
Herefordshire	0	-	-	1.3	1.3	2.2	0	0	3.5
S[illegible]pshire	4.5	-	-	7.4	2.9	7.9	58.1	0	73.4
Staffordshire	15.0	21.1	5.0	60.9	19.8	40.1	88.1	0	189.1
Warwickshire	9.0	21.1	1.0	56.6	25.5	45.8	51.3	0	153.7
Worcestershire	1.5	10.5	0.2	20.9	8.7	15.6	16.5	0	53.0
Cambridgeshire	0	0.2	-	5.1	4.9	5.0	0	0	10.1
Isle of Ely	0.1	0.2	-	5.4	5.1	4.0	0	0	9.4
Huntingdonshire	0	-	-	0	-	6.1	14.3	0	20.4
Norfolk	0.1	0.5	-	6.9	6.3	18.7	20.3	0	45.9
Suffolk	0.1	1.4	1.9	9.9	6.5	16.7	43.5	0	70.1
London	0	23.1	9.8	70.5	37.6	123.6	291.3	0	485.4
Bedfordshire	0	-	1.8	15.0	13.2	9.5	7.6	0	32.1
Berkshire	0	1.2	-	4.2	3.0	13.4	9.6	0	27.2
Buckinghamshire	0.1	1.2	-	12.4	11.1	11.5	0	0	23.9
Hampshire	0.1	2.3	1.3	9.6	5.9	32.4	33.9	38.2	114.1
Essex	0.1	4.7	2.7	33.5	26.0	26.7	72.7	34.8	167.7
Hertfordshire	0.1	4.7	2.2	22.4	15.4	19.1	16.5	0	58.0
Kent	0	2.3	-	43.1	40.8	26.7	76.7	23.2	169.7
Oxfordshire	0.2	-	-	12.2	12.0	7.7	1.9	0	21.8
Surrey	0	4.7	0.9	8.4	2.8	19.1	0	0	27.5
S[illegible]ex	0	2.3	-	9.1	6.8	24.8	34.9	0	68.8
Gloucestershire	0.2	4.5	3.1	23.7	15.9	22.8	0	0	46.5
Somerset	0.1	0.9	0.2	6.0	4.8	14.7	29.2	0	49.9
Wiltshire	0.1	0.9	-	3.4	2.4	10.6	5.8	0	19.8
Dorset	0.1	0.9	-	7.5	6.5	7.3	16.5	0	31.3
Devon	0.1	0.9	0.3	7.8	6.5	18.8	30.1	0	56.7
Cornwall	0.1	0.9	0.3	2.4	1.1	7.3	7.6	0	17.3
Monmouthshire	66.8	2.7	0.8	73.0	2.7	13.5	46.6	0	133.1
Glamorgan	59.4	4.5	5.8	88.5	18.8	37.2	114.4	18.5	258.6
Carmarthenshire	7.4	0.9	-	8.8	0.5	4.5	16.5	0	29.8
Pembrokeshire	0	0.9	-	1.0	0.1	3.1	1.9	26.2	32.2
Caernarvonshire	7.4	-	-	7.9	0.5	5.9	0	0	13.8
Denbighshire	0	-	0.8	1.8	1.0	5.3	0	0	7.1
Flintshire	7.4	-	0.8	11.9	3.7	5.1	5.8	0	22.8
Cardingshire	0	-	-	0	-	1.5	0	0	1.5
Merioneth	0	-	-	0	-	0.7	0	0	0.7
Montgomeryshire	0	-	-	0	-	1.5	1.9	0	3.4
Brecknockshire	0	-	-	0	-	1.5	0	0	1.5
Radnor	0	-	-	0	-	0.7	0	0	0.7

Table 3 (cont.)

SULPHUR DIOXIDE OUTPUT 1970

June 1973 Revision

Thousand tons

Scotland County	IRON & STEEL MANUFACTURE	ENGINEERING	CHEMICAL INDUSTRY	TOTAL INDUSTRY	OTHER INDUSTRY	Domestic	Power Stations	Oil Refineries	Total
Aberdeen		1.9		2.3	0.4	6.0	14.3	0	22.6
Angus				2.3	2.3	5.7	14.3	0	22.3
Argyll				0.1	0.1	1.2	0	0	1.3
Ayr				0.5	0.5	7.0	14.3	0.2	22.0
Banff				0.1	0.1	1.0	0	0	1.1
Berwick				0	-	0.4	0	0	0.4
Bute				0	-	0.4	0	0	0.4
Caithness				0	-	0.5	0	0	0.5
Clackman				0.1	0.1	0.9	0	0	1.0
Dumfries				0.1	0.1	1.8	0	0	1.9
Dunbarton				0.3	0.3	3.7	0	0	4.0
E. Lothian	1.2			1.3	0.1	1.1	0	0	2.4
M. Lothian		14.5		15.4	0.9	11.9	0	0	27.3
W. Lothian	6.1			6.2	0.1	1.9	0	11.4	19.5
Fife				1.7	1.7	6.6	7.1	0	15.4
Inverness				0.1	0.1	1.7	0	0	1.8
Kincardine				0.1	0.1	1.0	0	0	1.1
Kinross				0	-	0.1	0	0	0.1
Kirkudbright				0.1	0.1	0.6	0	0	0.7
Lanark	46.3	72.5		121.2	2.4	33.1	143.0	0	297.3
Moray				0.1	0.1	0.1	0	0	0.2
Nairn				0	-	0.2	0	0	0.2
Orkney				0	-	0.4	0	0	0.4
Peebles				0	-	0.4	0	0	0.4
Perth				0.2	0.2	2.6	0	0	2.8
Renfrew	6.1	5.6		12.2	0.5	6.8	0	0	19.0
Ross				0.1	0.1	1.2	0	0	1.3
Roxburgh				0.1	0.1	0.9	0	0	1.0
Selkirk				0	-	0.4	3.6	0	4.0
Stirling				0.3	0.3	3.9	19.1	0	23.3
Sutherland				0	-	0.4	0	0	0.4
Wigtown				0	-	0.6	0	0	0.6
Shetland				0	-	0.4	0	0	0.4
Ireland									
Antrim & Belfast				44.6		18.1	58.2	11.6	132.5
Down (excl. Belfast)				0		3	0	0	3.0
Armagh				0		2.5	0	0	2.5
Tyrone				0		2.8	0	0	2.8
Londonderry				0		3.8	2.4	0	6.2
Fermanagh				0		1.3	0	0	1.3
Totals				1662.1		1310.4	2540.4	257.9	5770.8

DATE	General Stability of mixing layer	Surface layer stability over sea	Mean plume wind $\bar{V}$ normal to track (ms^{-1})	$\bar{V}/_G$	$\theta_G-\theta_p$	$V_s/_G$	$\theta_G-\theta_s$
1.10.71	slightly stable	stable*	10	0.90	26°	0.73	35°
22.10.71	near neutral	stable*	22	0.94	12°	0.39	46°
2.11.71	slightly stable	stable*	18	0.91	23°	0.36	40°
9. 8.73	slightly stable	stable	12	0.89	19°	0.60	38°
4. 9.73	near neutral	stable	7.9	1.00	10°	0.56	44°
7. 9.73	slightly stable	stable	11.8	0.89	17°	0.38	47°
8.10.73	slightly unstable	slightly unstable	6.2	1.00	9°	0.62+	22°

TABLE 4

$\bar{V}$ is the mean wind in the mixing layer weighted by the observed concentrations of SO_2; G is the surface geostrophic wind speed, V_s is the 10-metre surface wind, θ_G is the surface geostrophic wind direction, θp is the mean weighted wind direction in the layer and θ_s is the 10-metre wind direction. The ratios $\bar{V}/G$ and $(\theta_G-\theta_p)/(\theta_a-\theta_s)$ were held constant along the back trajectories.

*inferred from land ascents and sea temperatures

+inferred from aircraft measurements

TABLE 5 The basic sulphur dioxide and mixing layer data for the 7 flights.

SULPHUR DIOXIDE

LEVEL		DATE 1.10.71	22.10.71	2.11.71	9.8.73	4.9.73	7.9.73	8.10.73
6	h				1484*	1700*	1395*	1971
	C				0.4	5	0.2	1.1
	V				16	15.2	4.1	10.9
	F				6	82	0.8	12
5	h				1027	1381	1145	1515*
	C				5	15	0.1	0.9
	V				14.8	12.1	3.9	12.2
	F				77	179	0.4	11
4	h	1333*		1140*	792	1089	897	1181
	C	3		0	9	30	0.1	0.8
	V	11.2		20.8	12.9	9.8	4.4	11.6
	F	37		0	120	291	0.4	9
3	h	927	878*	472	566	784	655	828
	C	3	6	19	13	31	0.1	2
	V	12	29.8	22.8	12.6	7.9	4.1	9.6
	F	35	170	440	165	244	0.4	15
2	h	547	392	302	330	472	412	491
	C	23	16	27	12	31	2	6
	V	10.1	24.7	20.8	11.3	5.6	8.1	7.9
	F	231	390	573	137	175	13	48
1	h	309	136	161	110	169	164	166
	C	37	18	31	13	27	11	13
	V	10	20.9	17.9	8.8	3	12.2	4.1
	F	370	378	554	114	80	131	52
TOTAL FLUX (tonne/hr)		122	112	193	85	187	32	27
MEAN VELOCITY		10	22	20	12	8	12	6

*1st Level above mixing layer

h = height (metres)

C = concentration of SO_2 in $\mu g/m^3$

V = mean velocity in ms^{-1} normal to track. ($1 ms^{-1} = 3.6 kmh^{-1}$)

F = flux in $g/m^3 \times m/s$.

TABLE 6

THE SULPHUR BUDGET		DATE						
SYMBOL	MEANING	1.10.71	22.10.71	2.11.71	9.8.73	4.9.73	7.9.73	8.10.73
h	depth of mixing layer (metres)	1050	450	600	1200	1700	1200	1600
V	mean speed of sampled air (ms^{-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 (hours)	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 (th^{-1})	238	158	370	149	320	67	78
SULPHUR DIOXIDE								
F_2	measured SO_2 flux on sampling track (th^{-1})	122	112	193	85	187	32	27
F_2/E_2	fraction of emitted SO_2 remaining off E. Coast	0.51	0.71	0.52	0.57	0.58	0.48	0.34
v_d (	deduced velocity of dry deposition (cms^{-1})	-	-	-	0.8	0.8	0.6	-
v_d (	assumed v_d implied by comparison (cms^{-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 (μgm^{-3})	30	15	25	10	22	6	7
SULPHATE (converted to equivalent SO_2)								
F_4 (	measured sulphate flux on sampling track (th^{-1})	-	-	-	18	30	13	6
F_4 (	deduced sulphate flux on sampling track (th^{-1})	38	14	63	-	-	-	-
F_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	($P_4 \equiv F_4 + R_4$)	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 (Fig7)	0.15	0.08	0.15	0.15	0.09	0.21	0.33
TOTAL BUDGET								
$100\,[F_2 + D_2 + P_4]/E_2 \equiv 100$		100	100	100	100	100	100	100
$100\,[F_2 + D_2 + P_4^*]/E_2$		99	99	98	103	100	101	99

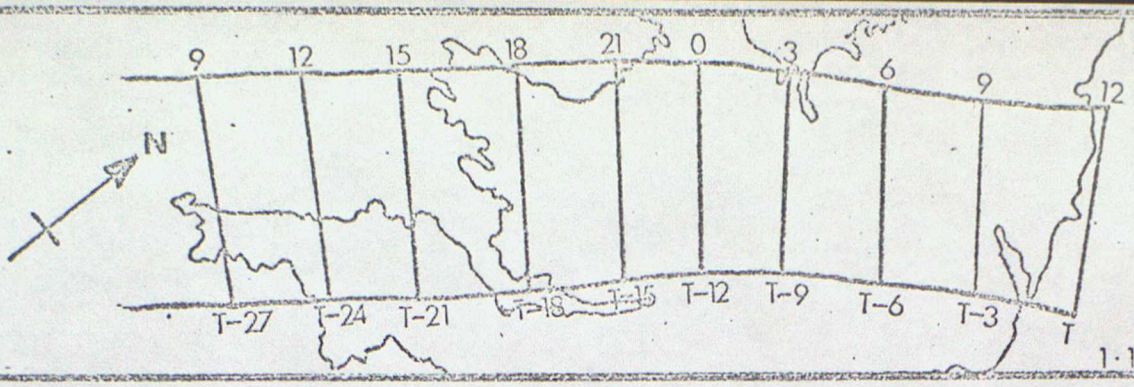
Notes: (a) all fluxes are measured in metric tons per hour.

(b) the basic known emission and flux data are E_2, F_2 and (in 1973 only) F_4. All other items are deduced from budget requirements and (in 1971 and on 8.10.73) by taking the assumed value of .

(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.

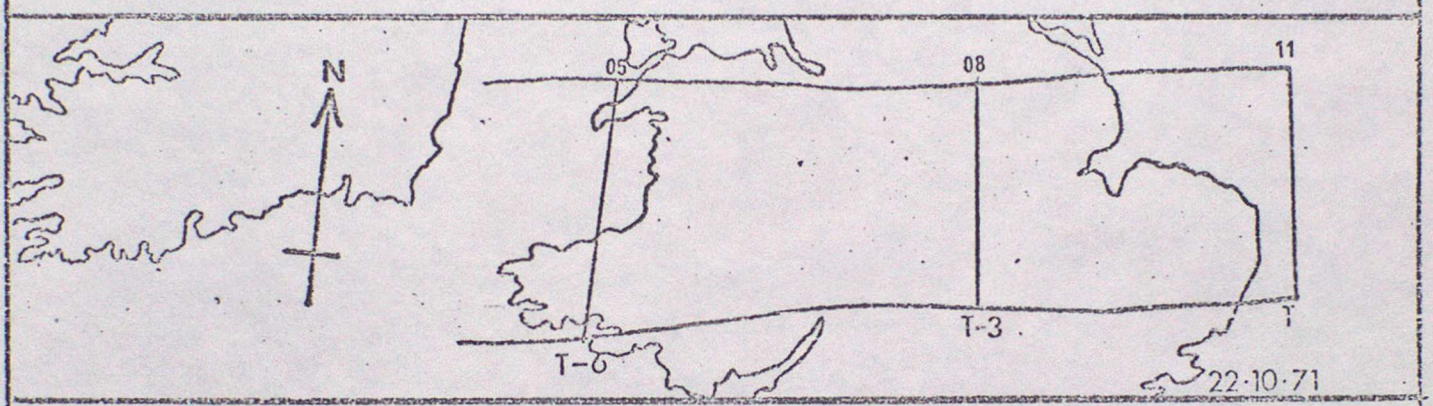
Distance downwind from source x (km)	$\frac{v_d}{V}$ ($\times 10^{-3}$)	Developing-plume model ($h \gg \sigma_z(x)$) Initial value of σ_z at source (metres)				Box model height h (metres)		
		10	20	30	50	500	1000	1500
90	0.5	84	85	86	86	91	96	97
	0.67	80	81	82	82	89	94	96
$\sigma_z(90) = 645$m	1	71	73	74	75	84	91	94
	2	51	53	54	56	70	84	89
125	0.5	83	84	84	85	88	94	96
	0.67	78	79	80	81	85	92	95
$\sigma_z(125) = 750$m	1	69	70	71	72	78	88	92
	2	47	49	50	52	61	78	85
160	0.5	81	82	83	83	85	92	95
	0.67	76	77	77	78	81	90	93
$\sigma_z(160) = 840$m	1	66	67	68	69	73	85	90
	2	44	45	46	48	53	73	81
200	0.5	79	80	81	81	82	91	94
	0.67	73	74	75	76	77	88	92
$\sigma_z(200) = 960$m	1	63	64	65	66	67	82	88
	2	41	42	42	44	45	67	77
250	0.5	77	78	78	79	78	88	92
	0.67	71	72	72	73	72	85	89
$\sigma_z(250) = 1100$m	1	60	61	61	63	61	78	85
	2	36	38	38	40	37	61	72

TABLE 7 Percentages of SO_2 remaining in the atmosphere as a function of distance downwind x, the ratio of velocity of dry deposition v_d to wind velocity V, and model. The developing-plume model assumes the sulphur dioxide is initially mixed through some depth at the ground and thereafter diffuses vertically according to Smith's $\sigma_z(x)$ curve for neutral stability, unlimited by any overhead inversion. The box-model assumes the sulphur dioxide is uniformly mixed through the mixing layer, which is capped by a strong inversion at height h.



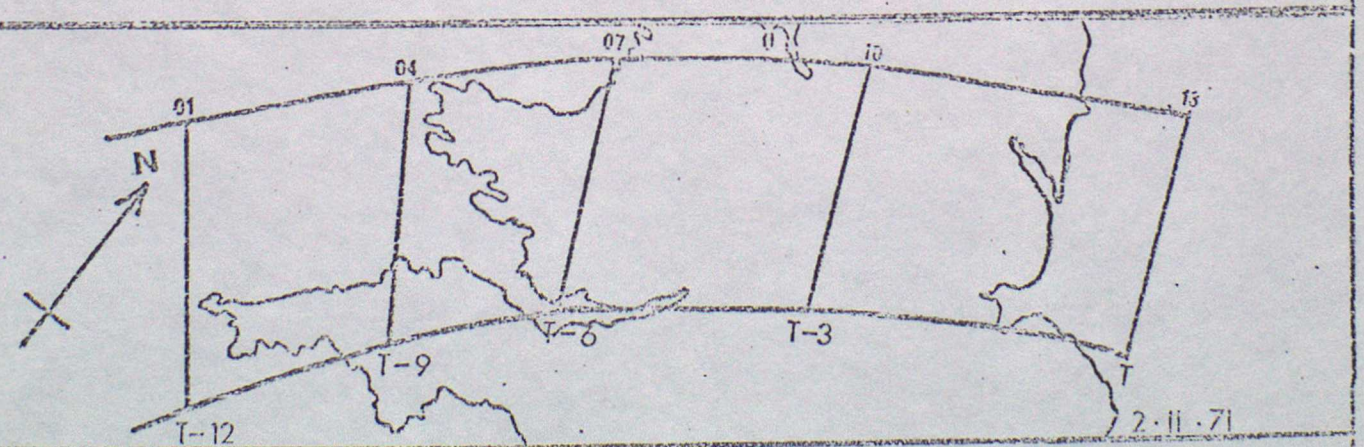
Wind : 220° - 250°, 6-8 ms^{-1}
Extensive night fog over Midlands.
Temp. Max (midday) 20°C.

Dry except for negligible light rain in Midlands.
Cloud : low stratus at 300m., 3 oktas St Cu at 1200m.
Mixing layer (midday) 1000m.



Wind : 260° - 270°, 20-23 ms^{-1}
Good visibility.
Temp : high for October, Max 18°C.

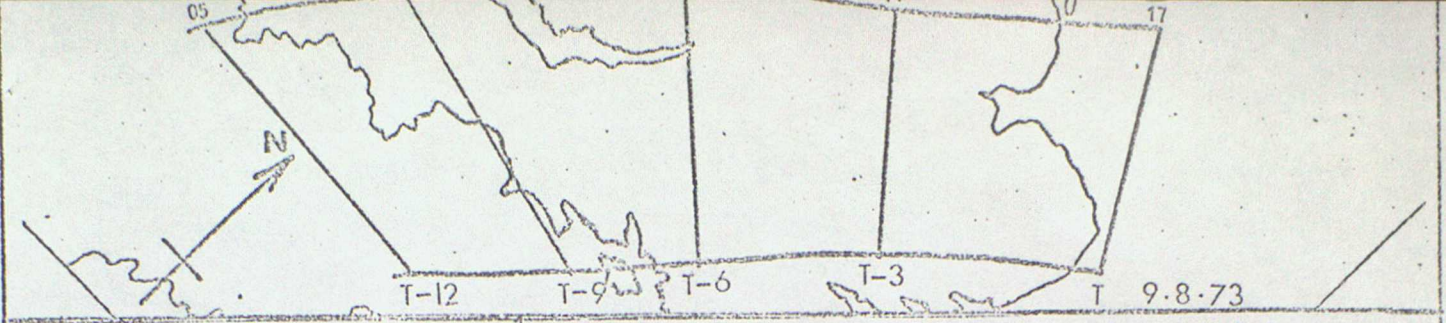
Dry, except for some light rain on Welsh coast.
Generally 3 oktas or less Cu & St Cu.
Mixing layer topped by marked inversion at 400m.



Wind : 240° - 260°, 15-22 ms^{-1}
Visibility moderate.
Temp. Max 15°C.

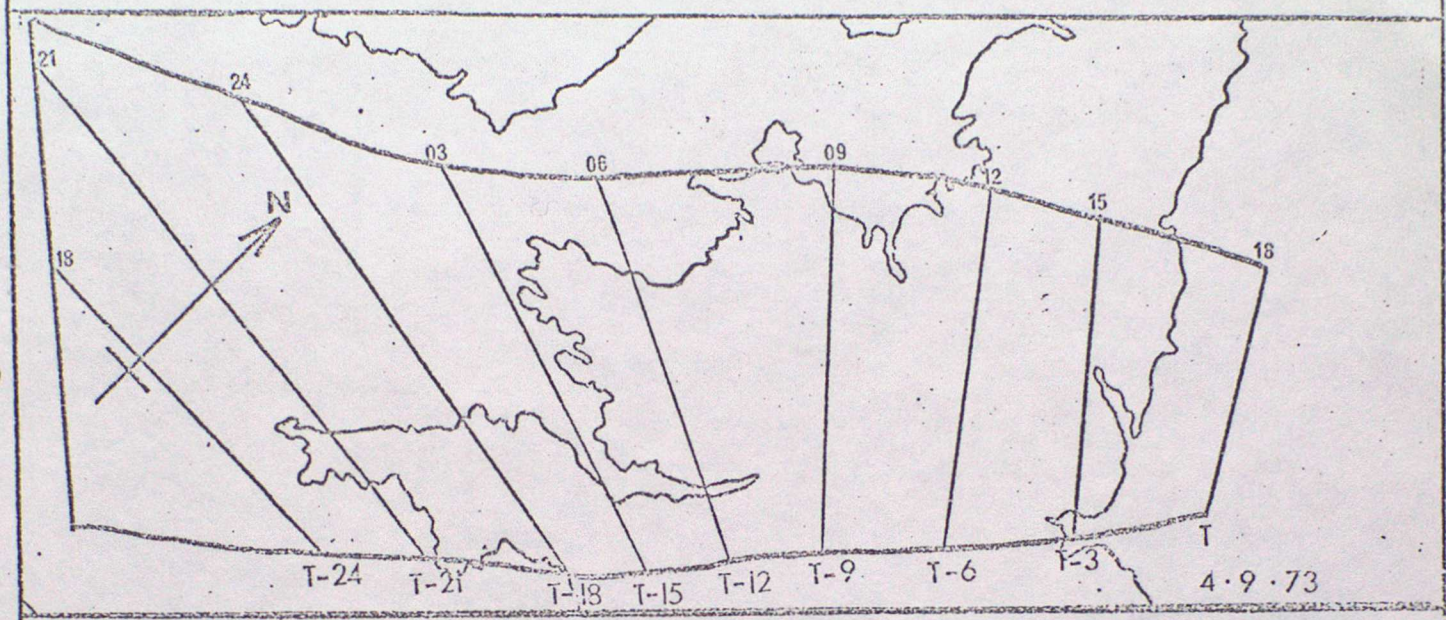
Dry.
Cloud : generally 2 oktas Cu & St Cu at 500m.
Mixing layer topped by marked inversion at 600m.

Figure 1



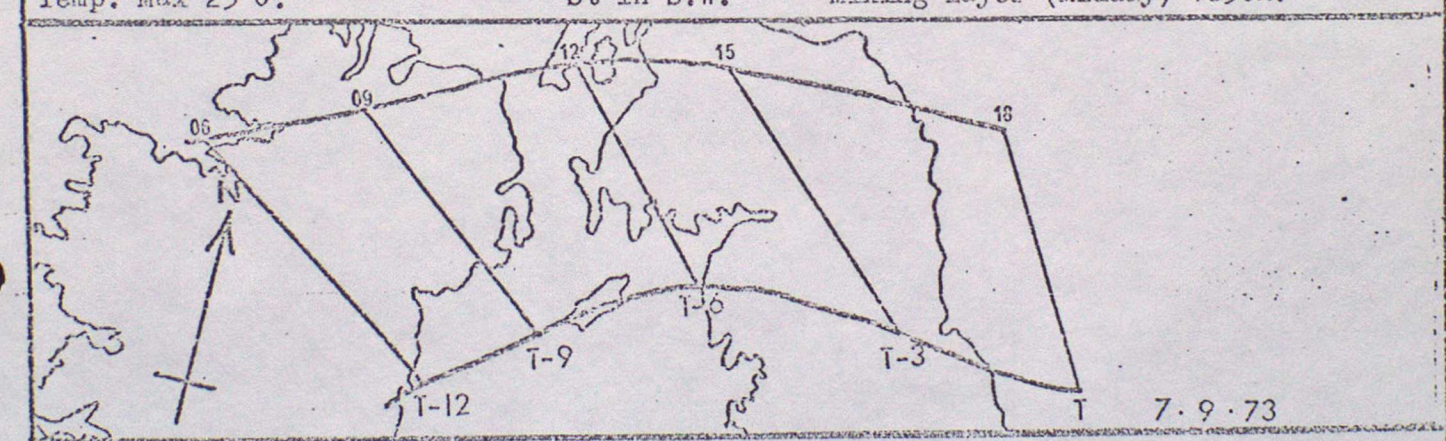
Wind : 230° - 240°, 13-20 ms^{-1}
Patches of fog in S.W. England.
Temp. Max 21°C.

Generally dry, some light rain on Welsh coast.
Cloud : mainly 4-7 oktas St Cu. at about 1000m.
Mixing layer (midday) 1200m.



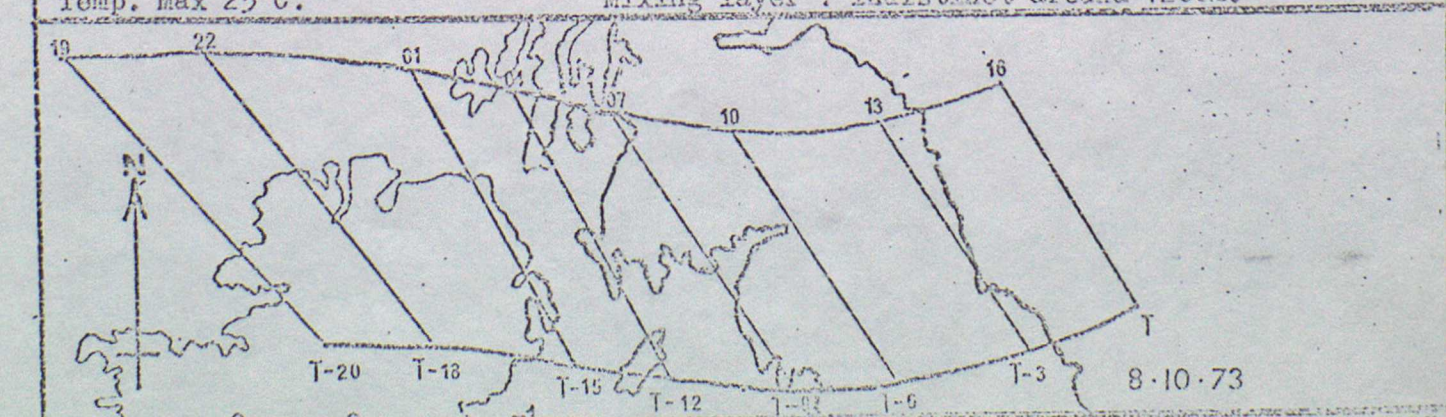
Wind : 220° - 230°, 5-9 ms^{-1}
Some fog in S.W. England.
Temp. Max 25°C.

Fine and dry except for local drizzle in S.W. England.
Cloud : small amounts of Cu & St Cu, except for 8 okts St in S.W.
Mixing layer (midday) 1850m.



Wind : 250° - 280°, 12-15 ms^{-1}
Visibility moderate to good.
Temp. Max 25°C.

Dry except for some light rain over western hills.
Cloud : Cu and St Cu. 500-1000m.
Mixing layer : indistinct around 1200m.



Wind : 260° - 280°, 5-10 ms
Mod. to good visibility.
Temp. Max 16°C

Warm front from N Ireland to the Wash gave some rain continuous and heavy along E coast & sampling track.
4-8 oktas St Cu at 700m. Mixing layer (midday) 1500m

Figure 3

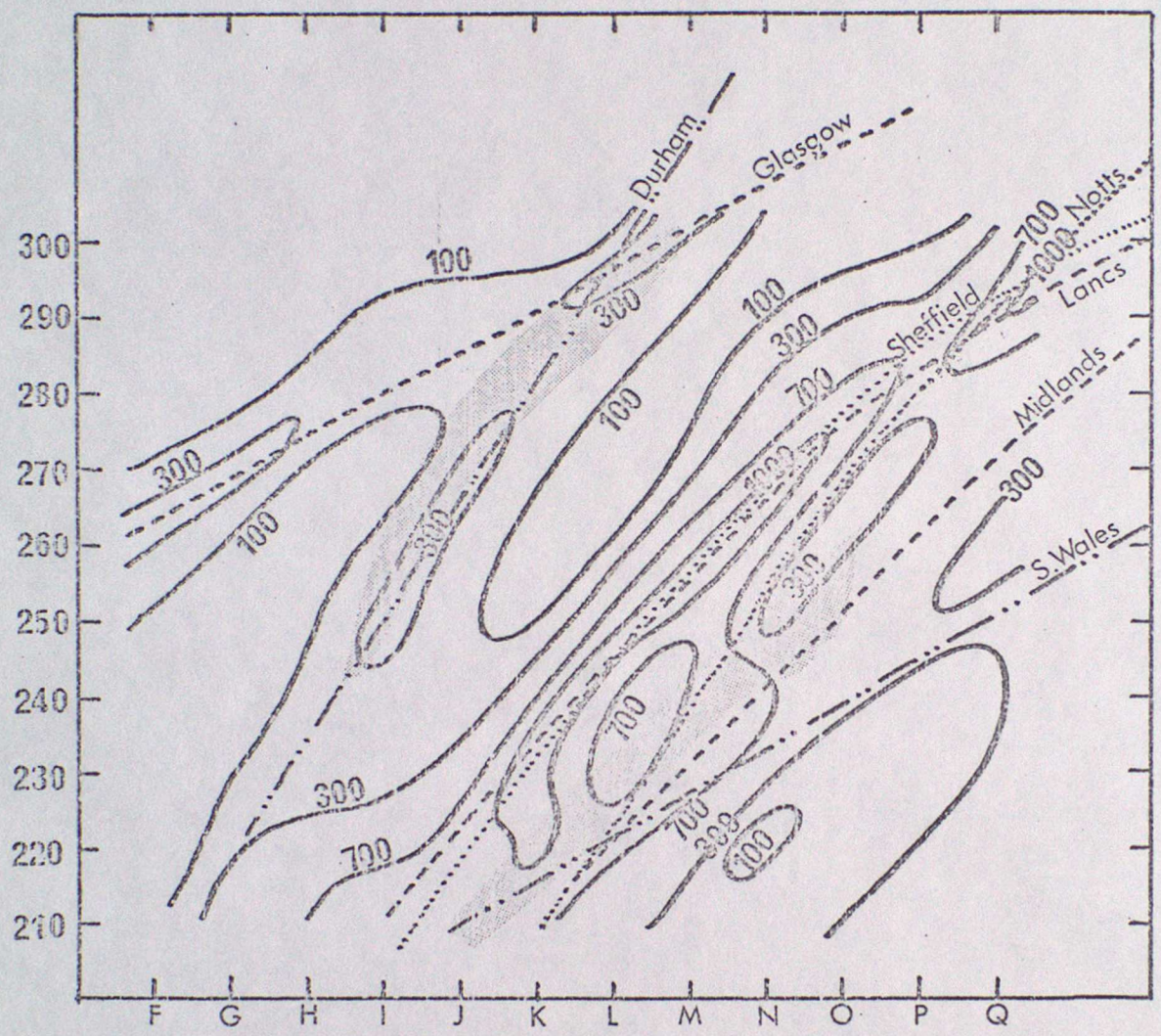

300
290
280
270
260
250
240
230
220
210
F
G
H
I
J
K
L
M
N
O
P
Q
Durham
Glasgow
Notts
Lancs
Sheffield
Midlands
S.Wales
100
300
700
1000

Figure 4

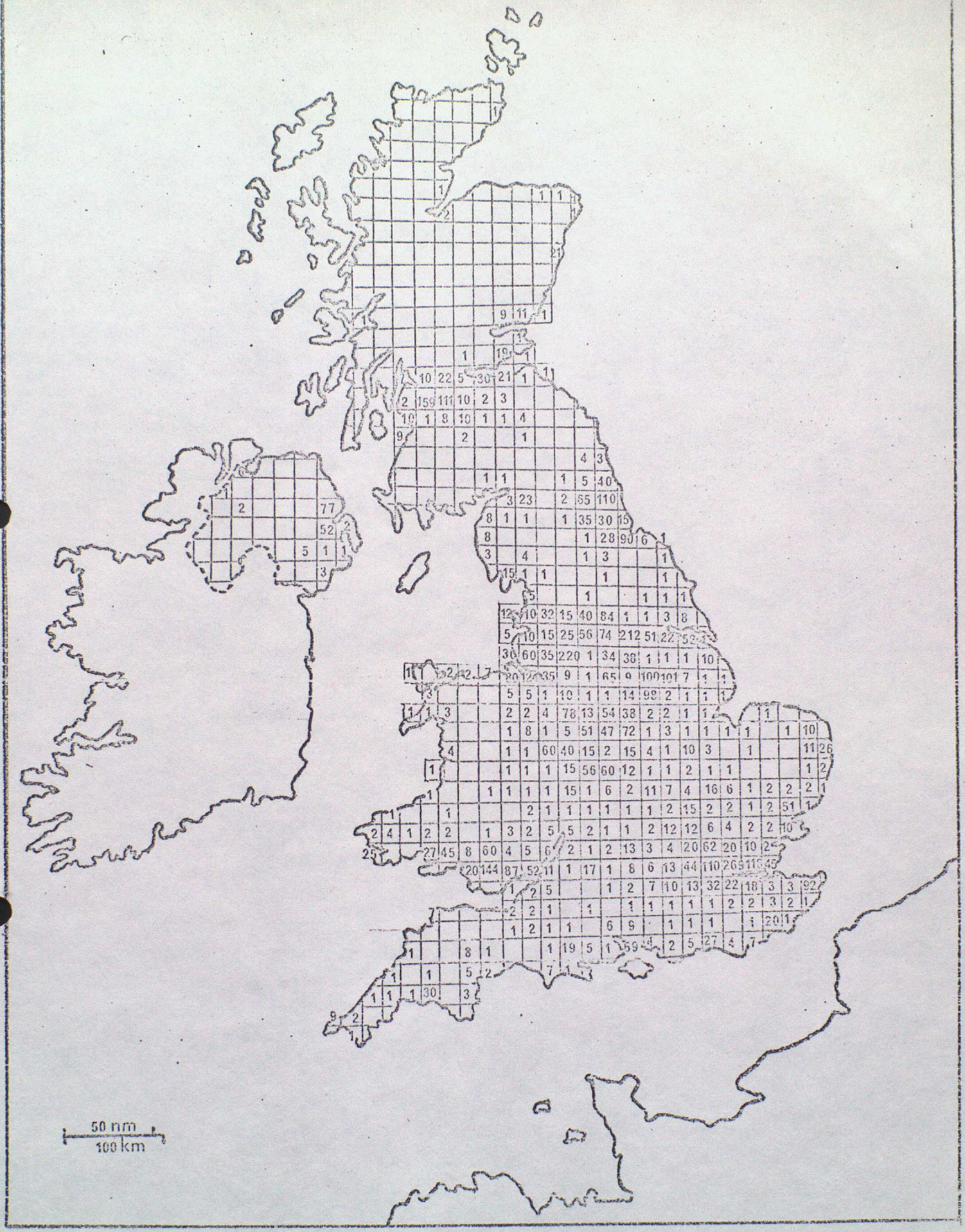
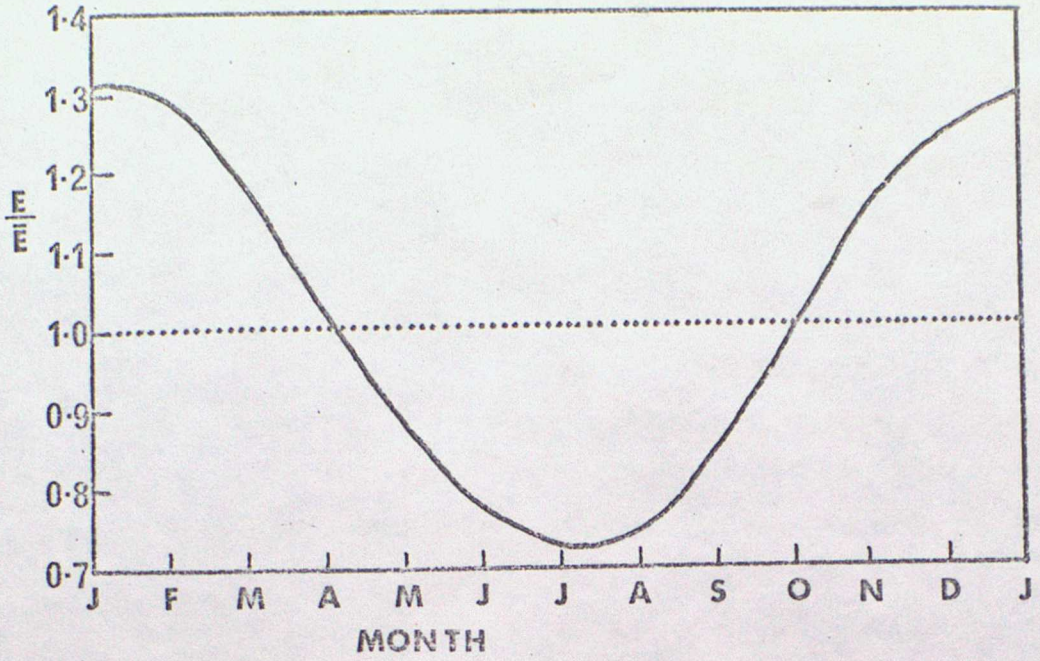
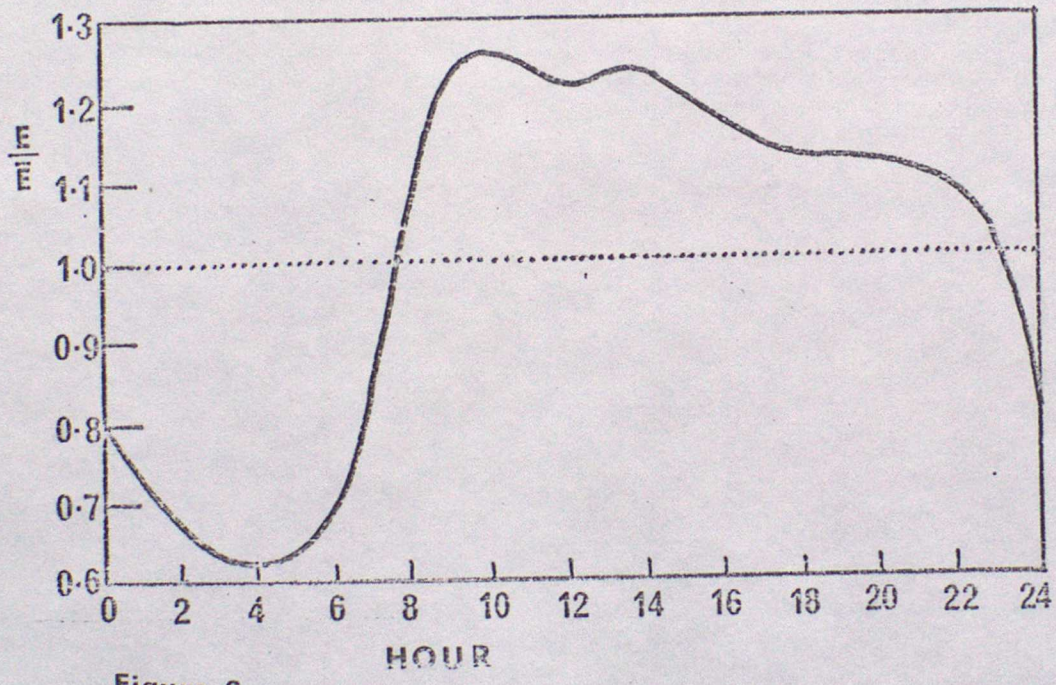


Figure 5. United Kingdom sulphur dioxide emissions for 1970 in thousands of tons per year.

Each square represents an Ordnance Survey 20x20 km region.


1·4
1·3
1·2
1·1
1·0
0·9
0·8
0·7
E/Ē
J F M A M J J A S O N D J
MONTH


1·3
1·2
1·1
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0·7
0·6
E/Ē
0 2 4 6 8 10 12 14 16 18 20 22 24
HOUR

Figure 6

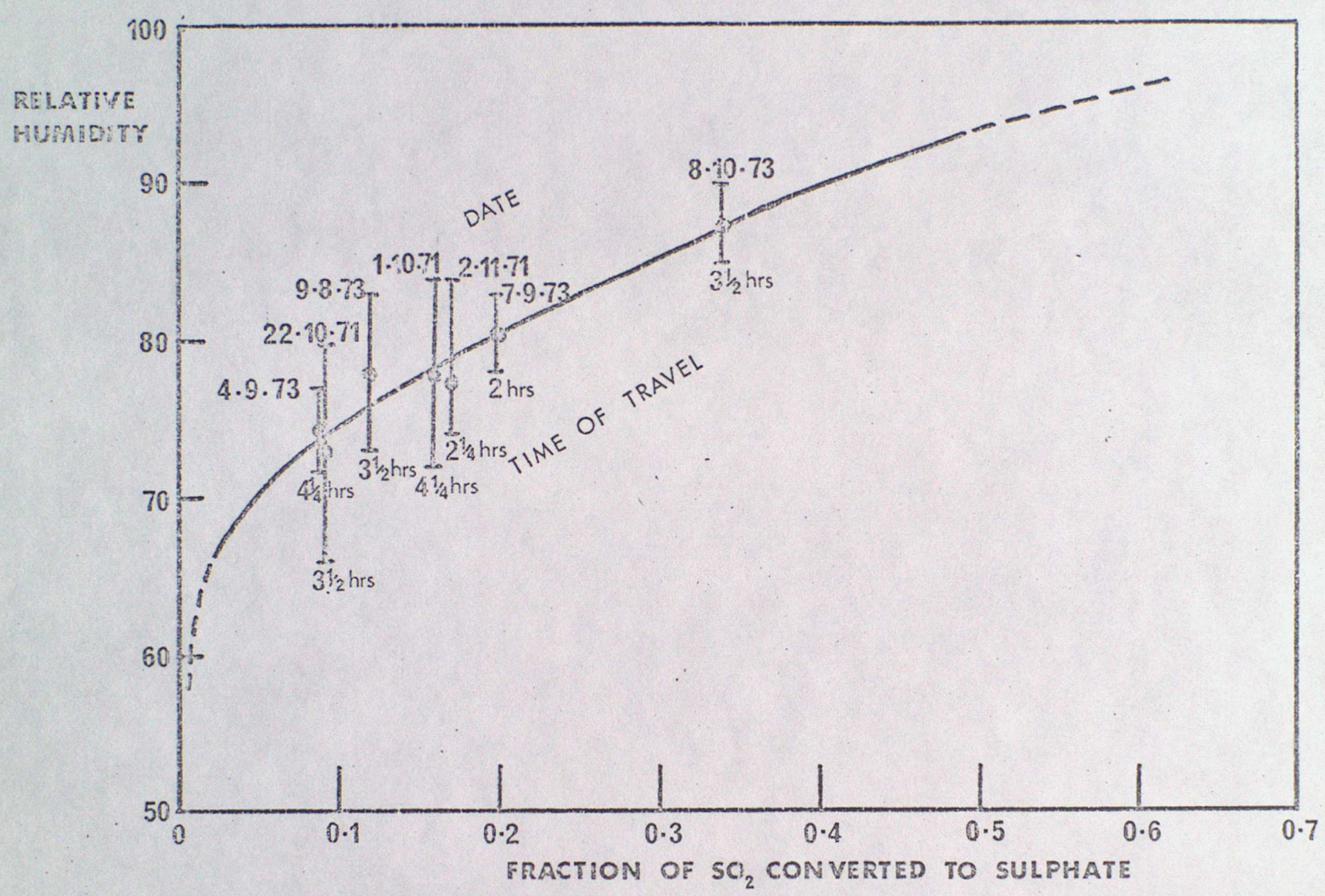
100
RELATIVE HUMIDITY
90
80
70
60
50
DATE
8·10·73
1·10·71
2·11·71
9·8·73
7·9·73
22·10·71
4·9·73
TIME OF TRAVEL
3½ hrs
2 hrs
2¼ hrs
3½ hrs
4¼ hrs
4¼ hrs
3½ hrs
0
0·1
0·2
0·3
0·4
0·5
0·6
0·7
FRACTION OF SO2 CONVERTED TO SULPHATE

Figure 7