# Deposition and transformation rates of sulphur oxides during atmospheric transport over the Atlantic

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ABSTRACT

Deposition and transformation rates are basic parameters for the determination of dispersion and deposition of aerosols. On the basis of an air trajectory model and appropriate aerosol measurements at the Faroes and the British Isles the concentration difference resulting from about 1 000 kilometres transport over water is used to determine an eddy deposition rate for sulphur dioxide and sulphate which corresponds to a deposition velocity of 2 cm/sec ±50 % and 0.4 cm/sec ±50 %, respectively. The transformation rate for sulphur dioxide to sulphate is found to be close to the eddy deposition rate for sulphate with a half life of 60 hours ±20 %. The clean Atlantic air contains 0.8 $\mu g/m^3$ ±50 % of sulphate and 0.2 $\mu g/m^3$ ±50 % of sulphur dioxide. These concentration levels are two to three times less than that previously assumed for global tropospheric averages used in estimating the global sulphur cycle, indicating that the natural production of sulphur at the free sea surface has previously been overestimated. Electron microscopic determination of the aerosol particle morphology and chemistry provides a unique and direct verification of the models used.

## 1. Introduction

The atmosphere transports pollutants between the countries in Western Europe. This causes increased concentration of sulphur and acid in precipitation as shown by S. Oden (1968) and L. Granat (1971).

Since 1972 there have been considerable efforts to quantify the cross boundary transport of atmospheric pollutants,[2] such as acid rain, acid particles and sulphur dioxide. Ground based and aircraft measurements improve the knowledge of dispersion and decay rates of gases and particulates, and different atmospheric dispersion models are used to compute the deposition pattern over Western Europe. Accurate determination of deposition rates and of chemical transformation rates for sulphur oxides are one of the most important requirements for obtaining reliable estimates of the deposition pattern on a regional scale. Laboratory experiments and small scale outdoor experiments may not be representative of the atmospheric conditions such as turbulence, stratification and catalytic characteristics during transport over large distances from hundreds to thousands of kilometres. Measurements in real scale are usually influenced by local sources and by the difficulty in distinguishing between local and distant sources.

This paper describes an investigation in which these difficulties have been overcome. A Danish measurement station at the Faroe Islands gives a new possibility to determine the decrease in concentration of pollutants during the transport over a distance of 1 000 km from the British Isles. On the basis of these "large scale experiments", described in detail below, we estimate the deposition and transformation rates for sulphur oxides, as a deposition velocity for sulphur dioxide of $V_d = 2$ cm/sec ±50 % and for sulphate of $V_d = 0.4$ cm/sec ±50 % and the transformation rate for $SO_2$ to $SO_4^{--}$, $T_{\frac{1}{2}} =$ 60 hours ±20 %.

Our results should be compared with previ-

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[2] This work has mainly been organized by OECD and in Scandinavia by Nordforsk (The Scandinavian Council for Applied Research) through the Norwegian Institute for Air Research, as described by B. Ottar (1973).

Table 1. *Determination of dry deposition velocity for sulphur dioxide*

| Year | Component | Reporter | Method | Typical horizontal scale (km) | Surface | Estimated average deposition velocity (cm/sec) |
|---|---|---|---|---|---|---|
| 1968 | $^{131}I$ | Van der Hoven | Review of experiments before 1968 | — | Water, grass<br>Snow, soil | 2<br>0.5 |
| 1971 | $SO_2$ | Liss | Theoretical | — | Water | 1 |
| 1972 | $SO_2$ | Spedding | Laboratory | — | Water | 0.4–1.4 |
| 1973 | $SO_2$ & $^{35}S$ | Garland et al. | Profile and tracer | 0.1 | Grass | 0.85 |
| 1974 | $SO_2$ | Fowler et al. | Profile | 0.2 | Wheat | 0.4 |
| 1974 | $SO_2$ | Garland et al. | Profile | 0.2 | Grass | 0.1–2.1 |
| 1974 | $^{35}S$ | Owens et al. | Tracer | 0.2 | Grass | 0.8 |
| 1974 | $SO_2$ | Shepherd | Profile | 0.4 | Grass | 0.8 |
| 1974 | $SO_2$ | Whelpdale | Profile | 2 | Snow, grass, water | 0.5–2.5 |
| 1975 | $SO_2$ | Eliassen et al. | Air trajectory (statistical) | 1 000 | Mixed | 2 |
| 1975 | $SO_2$ | Prahm et al. (this study) | Air trajectory (ground based) | 1 000 | Water | 2 |
| 1975 | $SO_2$ | Smith et al. | Air trajectory (aircraft) | 450 | Water | 0.8 |

ous investigations (Table 1). The deposition and the transformation rates for sulphur dioxide have been estimated by Eliassen & Saltbones (1975) for atmospheric long range transport on the basis of emission data, air trajectories and ground based measurements from a large number of measurement stations; $V_d$ = about 2 cm/sec, and the transformation rate for sulphur dioxide to sulphate, which is one magnitude of order less.

Chamberlain (1960, 1966) estimated a deposition rate for submicron particles also to be one magnitude of order less than the gaseous deposition rate. Van der Hoven (1968) summarizes a number of experiments on dry deposition of submicron particles which give deposition velocities at 0.2 to 0.5 cm/sec under stable atmospheric conditions. The gravitational fall velocity is less than 0.01 cm/sec (Junge, 1963).

Dry deposition is caused by sedimentation, impaction and chemical reactions. The sedimentation is negligible for gases and submicron particles. Because the aerosol is transported to the ground by eddy diffusion where impaction and chemical absorption takes place, and the air to water transfer resistance can be ignored (Litz, 1971; Spedding, 1972), the dry deposition mechanism should be called eddy deposition.

The eddy deposition rate and the rate of transformation of sulphur dioxide to sulphate are determined below for atmospheric transport over the Atlantic, and measurements on the "clean" Atlantic air determine a reference value for the cleanest possible air arriving to Europe. The validity of the model and the accuracy of the measurements is verified by results from the simultaneous analysis of different ions, trace metals and the particle chemistry, morphology and number distribution of the aerosol at the Faroe Islands.

## 2. Measurement site

The station is situated at the Faroe Islands in the North Atlantic at 62° N, 07° W on a cliff at a height of 740 m, 400 km north of Scotland and 500 km southeast of Iceland. The shortest distance to Norway is 650 km. In a direction sector of nearly 90 degrees between directly southern and close to western direction there are no populated areas, houses or roads in a distance of more than 500 km. The largest town at the Faroe Islands is Torshavn at sea level 10 km to the southeast with a population of 10 000 inhabitants. The total population in a radius of several hundred kilometres is about 40 000. Industrial activities are low with no

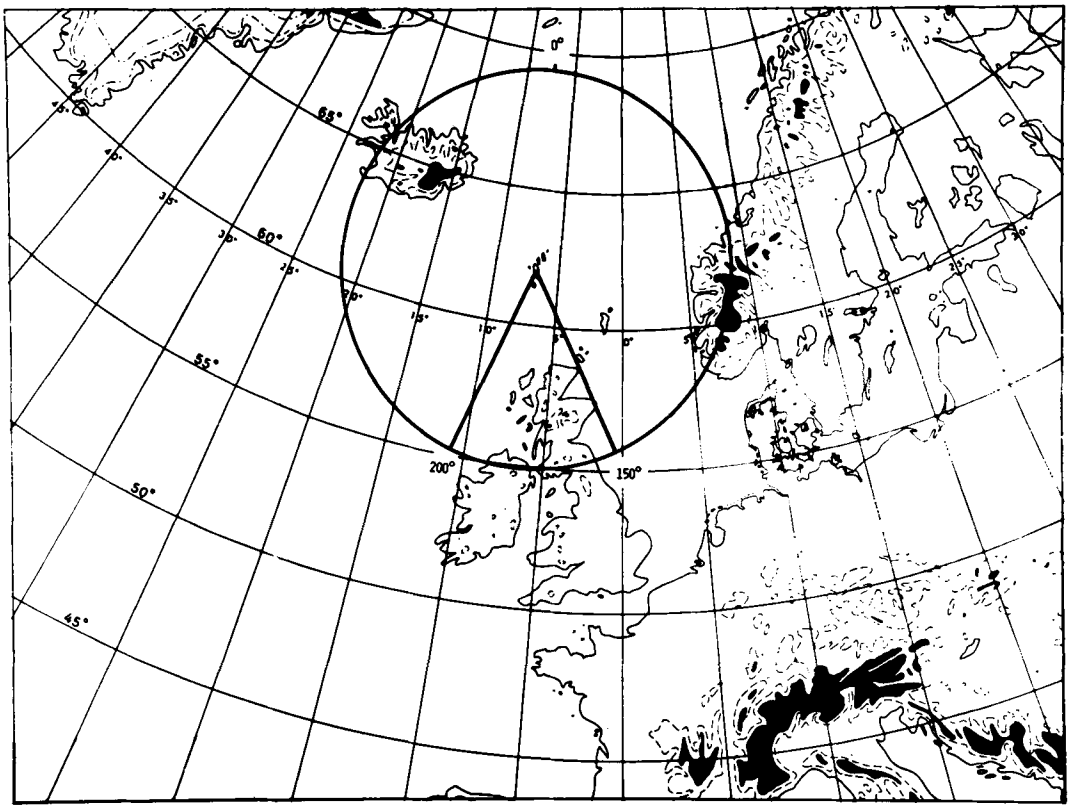


Fig. 1. Map showing the geographical area of the investigation. The circle has its centre at the measurement station at the Faroes and a radius of 900 km.

mining or heavy industry. A diesel engine exhaust 150 m below the measurement station in northeasterly direction does not influence the measurements substantially even with northeasterly wind, since the steep cliff prevents a significantly vertical wind velocity. Before passing the measurement station the air has usually moved thousands of kilometres above an unbroken ocean surface, giving a unique possibility for long time series of aerosol samples not contaminated by local sources. Although casually passing ships could function as local anthropogen pollution sources, a systematic meteorological analysis and extended series of 24 hours' sampling excludes this noise in the data. It is through a direction sector between south to west that the cleanest possible air is expected, where all local sources are excluded. Diurnal measurements of sulphate on particles during the period from November 1972 to June 1973 reported earlier (Prahm et al., 1974) show that no local sources substantially influence the sampling.

## 3. Sampling equipment

Quantitative determination of different aerosol components and measurements of the very low level of sulphur dioxide are obtained with a modified "OECD" instrument (Warren Spring Laboratory, 1966) supplemented with a high volume sampler pumping about 100 $m^3$ air per day through a series of two filters. A 50 mm Saturius 0.8 $\mu$ membrane filter is used to collect aerosol particles, followed by a 50 mm $NaHCO_3$ impregnated Whatman 40 filter for sulphur dioxide absorption. To prevent the filters from becoming wet due to cloud droplets, the inlet tube is elongated to about 3 m with

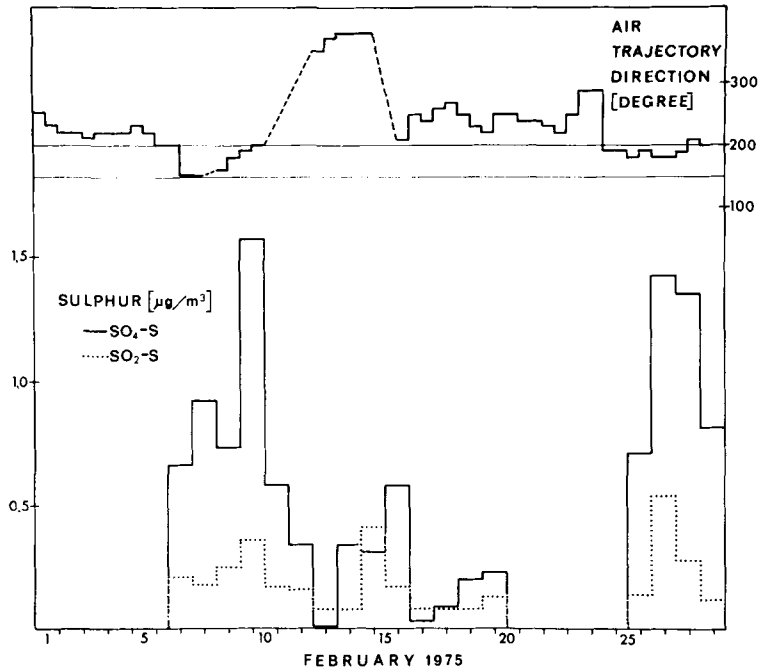


*Fig. 2.* The diurnal concentration of sulphur oxides measured as sulphur per cubic metre air volume and backward trajectory direction, computed twice a day. $SO_4$–S is corrected for seaspray.

a diameter of 50 mm. The loss in the tube is assumed to be negligible. To supply information on particle sizes, the station is equipped with an Anderson cascade impactor.

Aerosol concentrations in a height of about 90 m above sea level are measured by a high volume sampler at a neighbouring lighthouse. Since the concentration of sulphate from sea spray aerosol even in this height is larger than the anthropogen sulphate concentration, this data is not included. Daily precipitation was measured simultaneously with the aerosol sampling.

## 4. Meteorological analytical methods

Geostrophic air trajectories are computed to distinguish air masses which have passed over continental Europe and the British Isles from those having purely Atlantic trajectories during the 1–4 day period previous to measurement. The direction of the air flow is determined in the following way:

For every day in the period considered the direction sector is defined by the intersection point between a circle centred at the measurement station with a radius of 7.5° lat. (Fig. 1), and the back geostrophic air trajectory. Whenever the trajectory does not cut the circle, no direction is defined, and the air is considered to be of local origin. The important quantity is the relationship between the measured sulphur concentration and the measured direction sector.

The values from February 1975 are shown in Fig. 2. The lower graphs indicate the daily mean concentration of sulphur bound both as sulphate and as sulphur dioxide. The upper graph indicates the direction sector as defined above. When no direction sector could be defined the graph appears dashed. The direction expressed in degrees deviation from the north is indicated at the ordinate axis to the right. Air currents from the Western European industrial regions to the Faroe Islands mainly pass through the direction sectors between 150° to 200° and are found between the two horizontal lines in Fig. 2.

Geostrophic air trajectories are computed

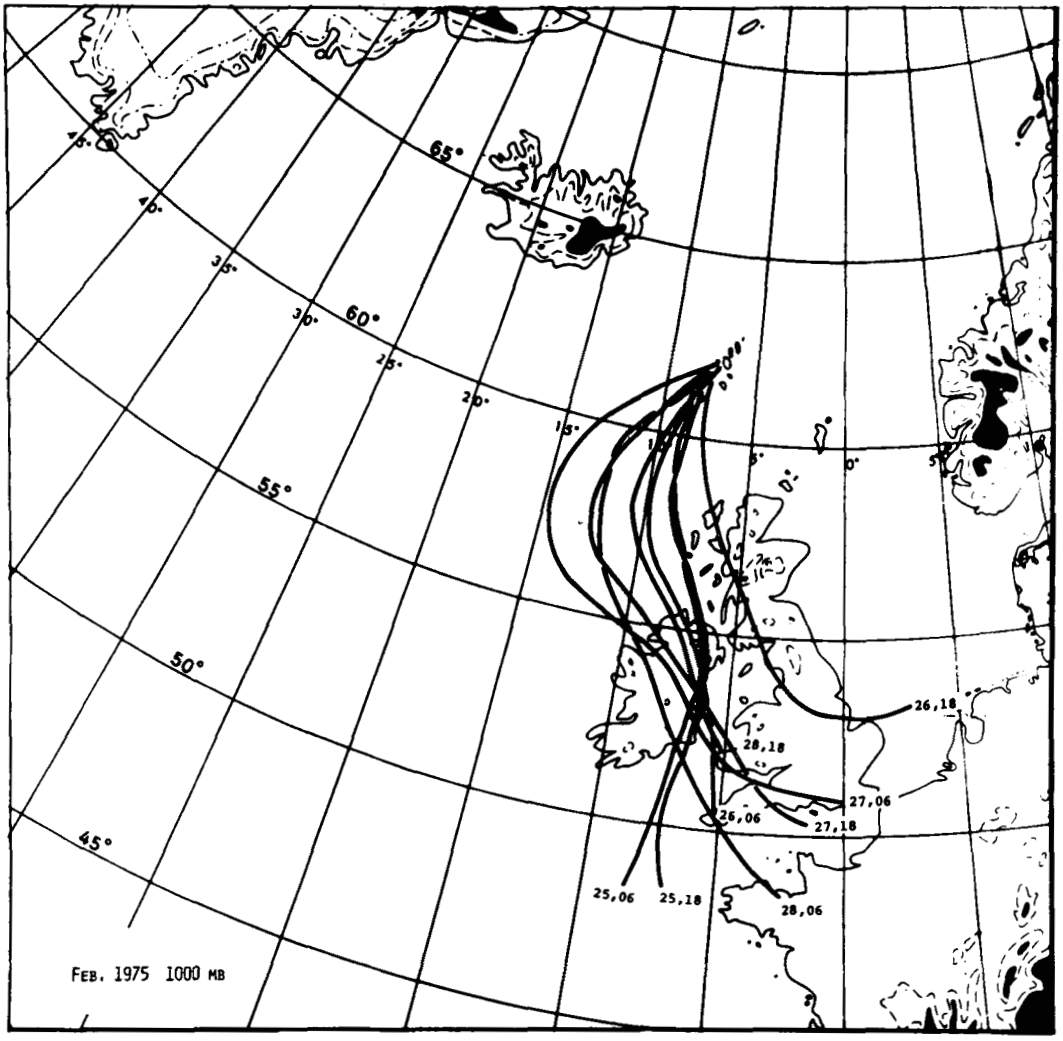


*Fig. 3.* Air trajectories during Long Range Transport episode in the 1 000 mb level. The first number at each trajectory gives the date and the second number gives the GMT time for arrival at the Faroes.

twice a day at 6 a.m. and 6 p.m., using the method of Petterssen (1956). The gradient of the pressure surface is computed on the basis of the nearest 16 grid points in the synoptic analysis mesh, with a distance of 300 km between the grid points.

## 5. Chemical analytical methods

After exposure the filters are transported in airtight polyethylene bags and then cut into one half and two quarters for use in wet chemical analysis, proton X-ray emission analysis (Folkman et al., 1974), and such special analysis as neutron activation (Heidorn et al., 1973) or scanning electron microscopy, respectively.

Detection limits for the proton induced X-ray emission analysis for the trace metals and bromine are less than 50 to 100 nanogram per filter or 0.5 to 1 nanogram per cubic metre air. For silicon it is about 20 ng/m$^3$ (Kemp, 1975). Analysis of sulphate as well as sulphur dioxide is performed by the spectrophotometric barium perclorate thorin method, with

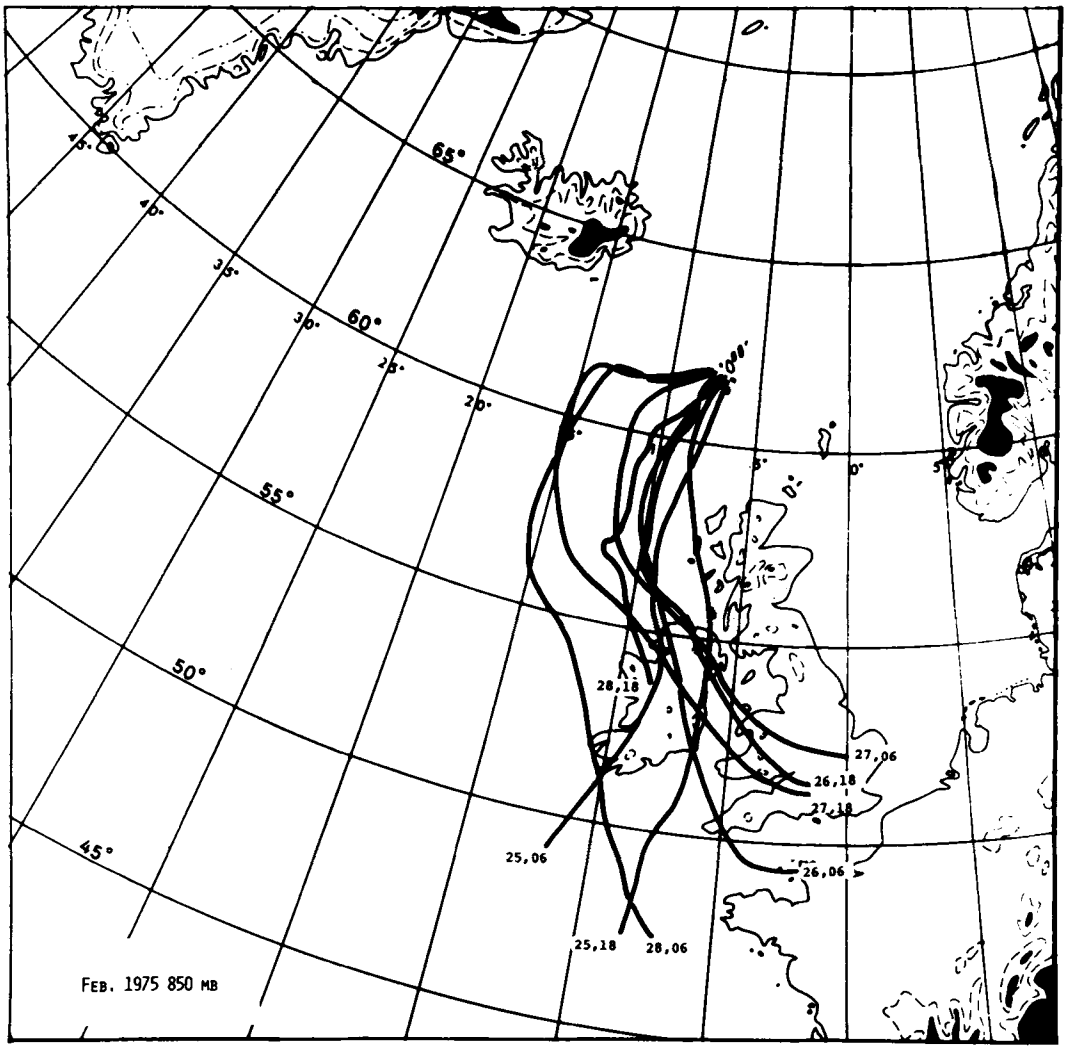


*Fig. 4.* Air trajectories during Long Range Transport episode in the 850 mb level, as in fig. 3.

a detection limit of about 1 to 3 microgram per filter, i.e. 10 to 30 ng/m³. $SO_2$ sampling efficiency is ensured by the high relative humidity at the sampling site. The amount of acid on particles is determined by a coloumetric titration using Gran's plot (Askne et al., 1973), having a detection limit of about 50 neq per filter or 0.5 neq per cubic metre air. Ammonium and nitrate are determined by a standard procedure using Nesslers reagent. Sodium is determined by using flame emission.

## 6. Chemical composition of the aerosol

The aerosol filters are changed every day at 12 GMT and labeled with the date of the sampling start. Sulphur concentrations during the month of February 1975 are shown in Fig. 2. Examination of the upper curve of trajectory direction shows that the days can be classified into four groups. From the 1st to the 6th and from the 20th to the 25th the apparatus was out of function. From the 6th to the 10th and

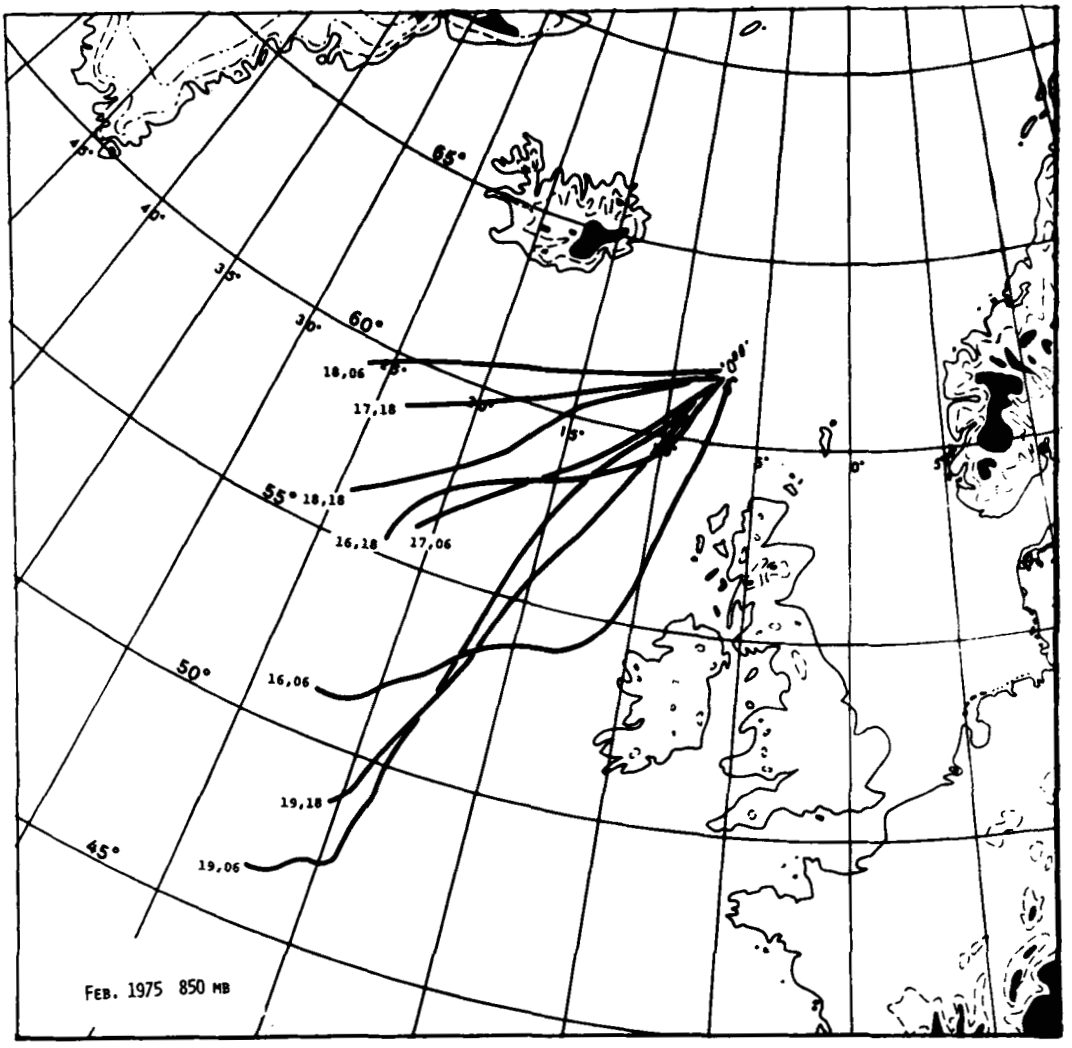


*Fig. 5.* Clean air backward air trajectories from February 1975 in the 850 mb level. The first number at each trajectory gives the date and the second number gives the GMT time for the arrival at the Faroes.

from the 25th to the 28th air passed through trajectory sectors between 150° to 200° and represents Long Range Transport (LRT) from the British Isles (Figs. 3 and 4). The period from the 10th to the 15th is influenced by air of "local origin". Between the 16th to the 19th is a period of clean Atlantic air (Fig. 5). The local wind observations in the periods from the 16th to the 19th and the 25th to the 28th were constantly in the south to west sector, so that no local sources contributed to the concentrations measured. The period with "local air" is weakly influenced by the sources at the Faroe Islands, and gives a sulphate level half as high as during the LRT episodes, while the sulphur dioxide is equivalent to the level during LRT.

The concentrations of sulphur and other chemical components during the clean air and the last LRT episode are given in Table 2. The sulphate values are corrected for sea spray on the basis of the sodium to sulphur concentration ratio in sea water. The sulphate and the sulphur dioxide levels are eight and three times higher, respectively, during LRT than in "clean

Table 2. *Aerosol and sulphur dioxide data, February 1975, Torshavn (nanogram per cubic metre)*

| | Source direction | | | | | | | | Atlantic average | British average |
|---|---|---|---|---|---|---|---|---|---|---|
| | Atlantic | | | | British Isles | | | | | |
| Date ... | 16 | 17 | 18 | 19 | 25 | 26 | 27 | 28 | 16 to 19 | 25 to 28 |
| $SO_2$–S | 80 | 80 | 80 | 130 | 140 | 540 | 280 | 120 | 90 | 270 |
| $SO_4^{--}$–$S_{cor.}$ | 30 | 90 | 200 | 230 | 710 | 1 420 | 1 350 | 810 | 140 | 1 070 |
| Sea–S | 190 | 90 | 90 | 90 | 30 | 70 | 80 | 110 | 115 | 70 |
| $NO_3^-$–N | 1.1 | 1.2 | 0.8 | 0.8 | 2.5 | 3.2 | 3.5 | 3.5 | 1 | 3 |
| $NH_4^+$–N | 8 | 19 | 15 | 10 | 29 | 28 | 42 | 40 | 13 | 35 |
| $H^+$ | 6 | 9.2 | 4.5 | 5.3 | 28.5 | 15.8 | 32.2 | 35.9 | 3 | 28 |
| Pb | 0.0 | 1.3 | 0.7 | 0.8 | 20.8 | 7.5 | 34.0 | 60.0 | 1 | 30 |
| Br | 5.4 | 2.1 | 2.4 | 2.2 | 3.6 | 2.1 | 3.5 | 7.5 | 3 | 4 |
| Zn | 0.7 | 2.1 | 0.0 | 0.4 | 126 | 55 | 209 | 391 | 1 | 19 |
| Cu | 0.4 | 0.5 | 0.0 | 0.0 | 0.7 | 0.3 | 1.1 | 1.5 | 0 | 1 |
| Fe | 3.7 | 7.9 | 0.0 | 0.0 | 47.6 | 26.5 | 57.7 | 96.1 | 4 | 57 |
| Si | 0.0 | 0.0 | 0.0 | 0.0 | 211 | 116 | 148 | 248 | 0 | 180 |
| Mn | 0.0 | 0.0 | 0.0 | 0.0 | 2.3 | 0.7 | 4.8 | 8.5 | 0 | 4 |
| V | 0.0 | 0.0 | 0.0 | 0.0 | 1.7 | 0.8 | 1.8 | 3.5 | 0 | 2 |

Atlantic air". The other analysed water soluble ions concentrations are raised by similar amounts (Table 2).

Both Atlantic and British aerosols are characterized predominantly by the presence of $NH_4^+$ and $SO_4^{--}$ ions. The ammonium to sulphate ratio is disproportionally low in the British air, because the neutralization of the additional sulphuric acid from British anthropogen $SO_2$ sources is limited by the low ammonia concentration associated with over-water transport (Brosset, 1975). The sulphur dioxide is oxidized by photochemical and catalytic reactions (Brosset et al., 1975; Atkins et al., 1972). About 30 nanoequivalents per cubic metre air of protons were measured during the LRT episode. This is more than the average level of 20 neq/m³ of protons at aerosol particles measured during a period of 100 days in a rural area in the southern part of Denmark, Keldsnor, close to large urban and industrial regions (Semb & Schaug, 1975; Prahm & Torp, 1975).

Those elements usually considered of continental origin, Fe, Mn, Si (soil dust) appear with relatively high concentrations during the LRT episode (Table 2). Also in agreement with our results are clean air aerosol particle concentrations of 4 ng/m³ iron as measured by Egorov et al. (1970) at Novaya Zemlya. Concentrations of lead are found to be 30 times higher during the LRT episode than in the clean Atlantic air. A clean oceanic air concentration of 1 ng/m³ lead was found as an upper limit over the Pacific by Chow et al. (1969), also in accordance with our results. Although bromine—as well as lead—is emitted through automobile gas exhaust, the bromine concentration is not raised during the transport episode. Earlier experiments show that 70% of the bromine has left the aerosol particles 18 hours after emission from an engine (Haar, 1971), most likely transferred to the gas phase through photochemical oxidation (Duce et al., 1975). The low bromine concentration that is measured is assumed to originate from the sea (J. L. Moyers et al., 1972). The particulate bromine concentration over the Pacific Ocean (Delany et al., 1973) of 5 ng/m³ is also in accordance with our values as well as with those of Moyers et al. (1972).

Vanadium concentrations determined by proton induced X-ray emission and by neutron activation analysis give the same average results for both the Atlantic and the British air (Table 2). The vanadium to lead ratio for the British air is similar to that given by Peirson et al. (1973). Our methods were not sensitive enough to determine whether the clean Atlantic air did carry vanadium of anthropogen origin in the concentration range between 0.1 and 1 ng/m³.

## 7. Washout ratio

The precipitation sampler, placed in the height of 700 m measures to some extent the cloud droplet concentration from the fog usually surrounding the cliff and is checked every day and analysed for strong acid, sulphate and sodium. The period with measurements in February 1975 (Fig. 2) included six days with rain. Usually no rain is falling during the long range transport episodes i.e. anticyclonic weather conditions. The washout ratio is defined as the ratio between the concentration in air per kilogramme air and the concentration in rain per kilogramme rain (Chamberlain, 1960). The washout ratio for sulphur was 4 000 and for sodium 24 000. This is 2 to 10 times the value given for sodium measured at the British Isles by Peirson et al. (1973). The higher washout ratio found in our study might to some extent be caused by our precipitation sampling technique, including both the dry and the wet deposition.

Table 3. *Nomenclature*

| | |
|---|---|
| $q(t)$ | concentration of $SO_2$ |
| $p(t)$ | concentration of $SO_4^{--}$ |
| $r(t)$ | concentration of trace metal |
| $h(t)$ | mixing height |
| $t$ | air travel time |
| $T$ | travel time between two measurement stations |
| $\tau$ | residence time |
| $T_{\frac{1}{2}i}$ | half life |
| $V_i$ | deposition velocity |
| $K_i$ | decay, transformation, and deposition rate |
| $i=1$ | decay for $SO_2$ |
| $i=2$ | deposition for $SO_2$ |
| $i=3$ | transformation for $SO_2 \rightarrow SO_4^{--}$ |
| $i=4$ | deposition for $SO_4^{--}$ |
| $i=5$ | deposition for trace metal |
| $\alpha$ | mixing height correction factor |
| $\beta$ | cross-wind dispersion correction factor |
| $\gamma$ | correction factor for $SO_2$ |
| $x$ | air volume travel distance |
| $L$ | air volume width |
| $\theta$ | angle cross-wind spread |
| $V_d$ | deposition velocity |

## 8. Aerosol particle distribution

Because of the favourable experimental conditions present at the Faroe site, it was felt that microscopic examination of the aerosol could provide direct evidence of differences of air mass trajectories as well as permit identification of the origin of chemical differences in the aerosol. Two filters were selected for scanning electron microscopy. The first day—19 February 1975—representing clean Atlantic air and the second day—26 February 1975—representing air transported from the British Isles. On both days the local wind directions were steady through the clean air sector described in chapter 2.

Filter segments from the two exposures were prepared simultaneously. Initial evaporation of 50–100 Å carbon (from arc) at below $10^{-5}$ torr in a series of short steps at continuously varying angles of attack on rapidly rotating filters, was followed by 50–100 Å of gold (tungsten filament) under the same conditions.

Using a jeol JSM-S3 scanning electron microscope, a series of photographs at magnifications from 300–30 000 were examined for particle size distribution. The smallest particles which could be identified had diameters in the range 200–400 Å (0.20–0.04 microns): The largest particles found on the filters were between 10 and 20 microns. Images at 10 000 could be used to count the distribution from 0.02–2.0 microns, while images at 1 000 could be used for particles from 0.8 microns upwards. Sufficient images were examined to provide a significant number of particles in each logarithmic size range between 0.02–20 microns (with the exception of the largest and smallest) measured with a hand held microscope. For the purpose of this preliminary exploratory study the total number of particles in each survey was limited to between 500 and 1 000 The relative confidence limit for each size range is therefore of the order of 50% with equal contributions to error coming from statistics, missed and twice counted particles, and overlap between the size ranges. The general behaviour of the distribution is not expected to change with a larger sample or with different counting methods.

The filter background is of such a dimension (1–2 microns) that there is no interference between filter texture and particles provided that an appropriate choice of magnification is made for each particle size range.

Counting is straightforward, except that the long range transport sample contains clusters 3–10 microns in diameter composed of small

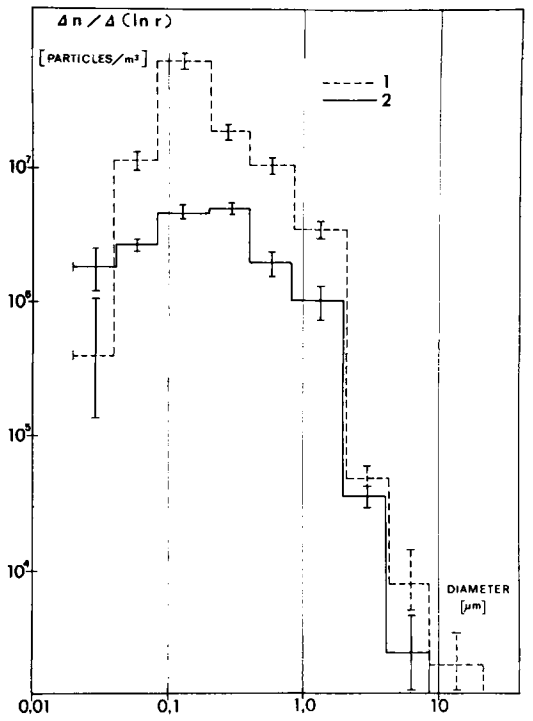


*Fig. 6a.* Optical diameter versus the number of aerosol particle distribution. Curve 1 on February 26 and curve 2 on February 19, 1975.

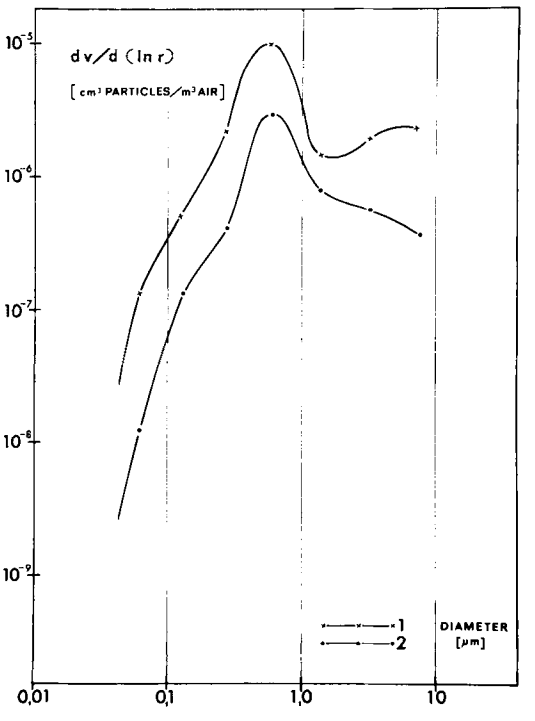


*Fig. 6b.* Optical diameter versus the volume of aerosol particle distribution. Curve 1 on February 26 and curve 2 on February 19, 1975.

particles of 0.1–0.5 microns in diameter. Monolithic particles of similar size are occasionally covered with small particles as well. The difficulty is to establish a systematic procedure for counting these agglomerates. It is not clear if they should be considered as one large particle or be divided up into their constituent particles. In the sample measured the results were not significantly altered by choice of procedure because of the relatively small number of large composites. If a significant fraction of the total mass is composed of such particles, then the number of small particles contained therein becomes important, and the choice of procedure would affect the ultimate form of the distribution. An additional question is the possibility of loss of aerosol from the surface of the filter towards the bulk by means of a diffusion or other transport mechanism, but it is felt that this effect will only become important on filters of significantly larger pore size than those used here: The maximum loss of particles does not exceed a factor of two.

Fig. 6*a* gives the optical diameter versus the number of aerosol particles, and Fig. 6*b* the optical diameter versus the volume of aerosol particles in a series of equal logarithmic size ranges from 0.02–20 microns. The distributions from the two days have almost the same shape, and most of the particles have a diameter from 0.05 $\mu$ to 5 $\mu$. Maximum for the distribution function is at a diameter of about 0.1 $\mu$. The number of particles is about 10 times higher on 26th February than on 19th February. Considering the height of the measurement station, the distributions do not deviate significantly from other measured clean air distribution (Junge, 1963; Junge & Jaenicke, 1971; Meszaros & Vissy, 1974). It is therefore assumed that the sampler inlet tube does not distort the distribution function.

Scanning electron microscope examination of the two filters shows that each of the two episodes is characterized by completely different particle morphologies consistent with the known trajectories. It was found that all detectable

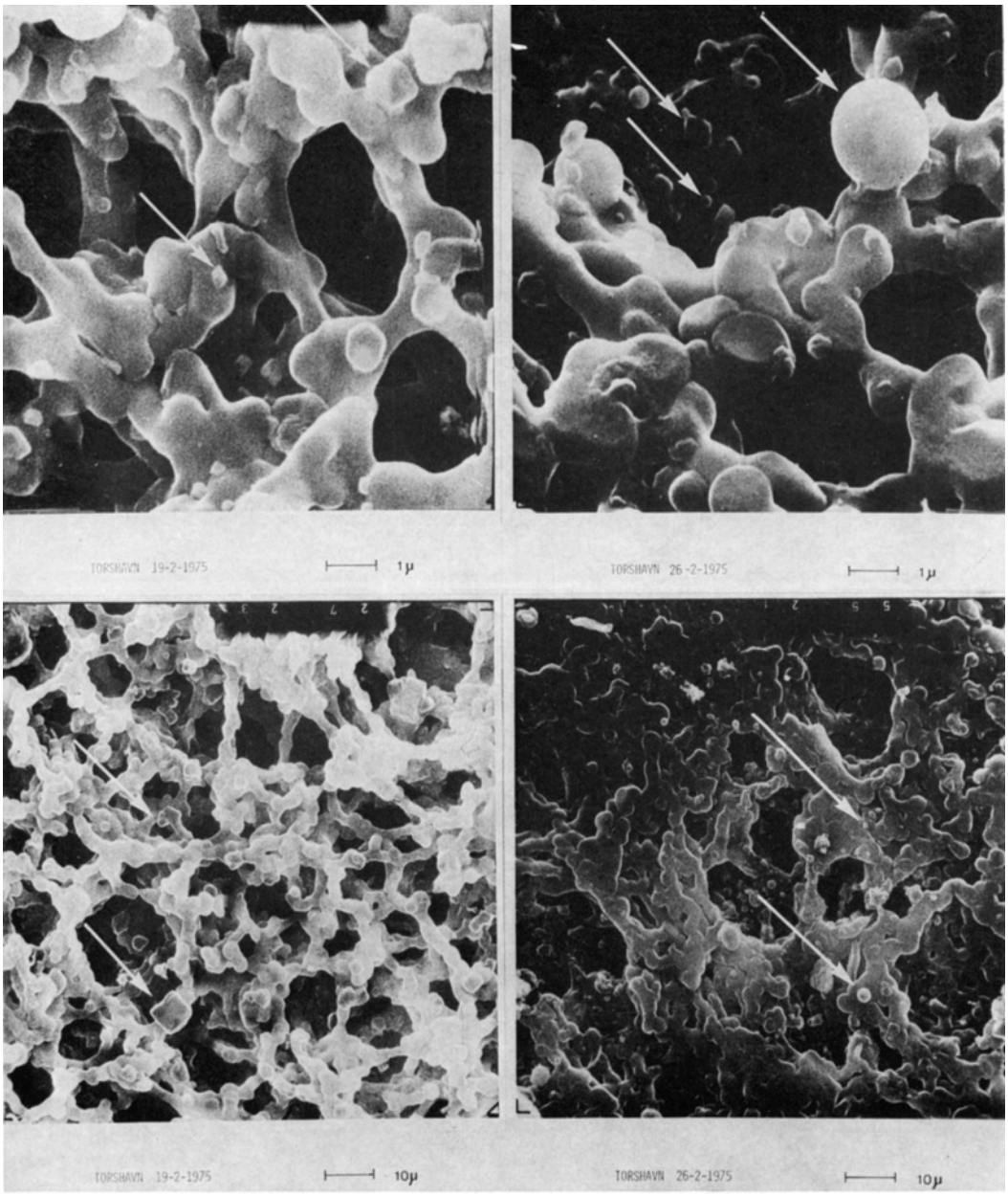


*Fig. 7.* SEM photographs of clean Atlantic aerosol (left) and Long Range Transport aerosol (right). The Atlantic air sample, February 19, 1975, shows crystalline particles only, The British air sample, February 26, 1975, contains both crystalline and spherical particles.

particles on the filter from 19th February were crystalline and most of them clean cubes (Fig. 7). The filter from 26th February (Fig. 7) contains particles of three distinct morphologies: Spheres, crystalline flakes and crystals with various rectangular cross-sections.

The SEM equipped with energy dispersive detection for X-ray analysis (EDAX) provides

Table 4. *Variations in relative elemental concentrations at aerosol particles as a function of particle morphology*

| No. | Filter sample | Particle shape | Particle size ($\mu$) | Uncorrected channel peak height | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | Na | Mg | Al | Si | S | Cl | K | Ca |
| 1 | Clean air | Cube | 1.5 × 1.5 | 2 | — | 1 | — | 6 | 20 | 2 | 4 |
| 2 | Clean air | Cube | 3.0 × 3.0 | 14 | — | — | 2 | 6 | 48 | — | — |
| 3 | LRT | Rough spherical agglomerate | 4.0 | — | — | 10 | 25 | 1 | — | 2 | — |
| 4 | LRT | Rough cube agglomerate | 2.0 × 2.5 | — | — | 6 | 21 | 1 | — | 2 | — |
| 5 | LRT | Rough flat flake agglomerate | 4.0 × 3.0 | — | 10 | — | 18 | 6 | — | — | 2 |
| 6 | LRT | Composite sphere | 1.6 | — | — | — | 55 | 3 | — | — | — |
| 7 | LRT | Smooth homogeneous rhomb | 1.0 × 1.0 | — | — | — | — | 30 | — | — | — |
| 8 | LRT | Smooth homogeneous sphere | 1.0 | — | — | — | 1 | 11 | — | — | 1 |
| 9 | LRT | Smooth homogeneous rod | 6.0 × 1.4 | — | — | — | 3 | 35 | — | — | 35 |

a unique opportunity for the determination of chemical composition of these aerosols on particle by particle basis. Table 4 lists some particle characteristics, as determined by the relative peak height for each element, found in a preliminary EDAX survey of the two aerosol samples discussed above. Because of variations in scattering and beam penetration, particles of different sizes and shapes are not directly comparable in terms of absolute chemistry, but variations in the relative elemental distribution from particle to particle can be determined by direct comparison of the elemental peak height ratios. Particle shapes 1 and 2 are typical cubes from the clean air sample and would appear to be crystals of sea salt. In addition to chlorine, the crystals contain magnesium and sulphur, probably originating from sea spray in accordance with the relatively high sodium contents (Table 1) and the air trajectory (Fig. 5). (Sodium is of too low atomic number to be detected.) The EDAX survey of the LRT aerosols shows that particles of each distinct morphology have similar chemistry. Particle shapes 3 to 6 are examples of typical agglomerates of different sizes and shapes, and contain relatively larger amounts of Si with little sulphur. A general survey of the filter indicates that the chemical composition of most agglomerates appears to be similar with respect to this large silicon to sulphur ratio. Particles shapes 7 to 9 are examples of the smooth, apparently homogeneous class of particles, which are all characterized by high sulphur contents. It is felt that these distinctive classes 4 to 6 and 7 to 9 represent sources to the aerosol from the ground (high silicon content) and from continuous gas phase reactions (high sulphur content).

Some of the rectangular shaped crystals had a high content of sulphur but no cloride or magnesium. This might be ammonium sulphate (Meszaros & Vissy, 1974) in combination with some other components. Some spheres contain only sulphur. The trace metals and sulphur at these spheres are probably of anthropogen origin in accordance with the high total contents of sulphur and trace metals (Table 2) and the air trajectory (Fig. 4). Measurements during the autumn 1974 with the Climet particle counter and the Anderson cascade impactor also show that the main distribution of particles and particle mass lies below 1 $\mu$ and 3 $\mu$, respectively.

## 9. Air trajectory box model

In the following discussion the decay of aerosol concentrations is described by a simple air

trajectory box model, using the parameters presented in Table 3. A Lagrangian computation is used to follow a certain air volume during advection along geostrophic air trajectories (Eliassen & Saltbones, 1975), and decay processes of deposition and chemical transformation are taken to be linear.

It is assumed that the aerosol is homogeneously mixed into the air volume at the time $t = 0$, and during transport vertical eddy diffusion maintains homogeneous mixing. The air volume advects over a homogeneous surface, e.g. the sea, and decay rates are assumed independent of time and concentration. The deposition rate might contain contributions from both wet and dry deposition. If the mixing height is constant and the lateral diffusion negligible, the concentration for sulphur dioxide, sulphate and trace metal can be described by the following first order linear differential equations:

Trace metal: $$\frac{dr(t)}{dt} = -K_5 r(t)$$

$SO_2$: $$\frac{dq(t)}{dt} = -K_1 q(t)$$

$SO_4$: $$\frac{dp(t)}{dt} = -K_4 p(t) + K_3 q(t)$$

All parameters are explained in Table 3.

The solutions for these equations are:

$$r(t) = r(t=0) \exp(-K_5 t) \tag{1}$$

$$q(t) = q(t=0) \exp(-K_1 t) \tag{2}$$

$$p(t) = p(t=0) \exp(-K_4 t) + \frac{K_3}{K_1 - K_4} q(t=0) \left(\exp(-K_4 t) - \exp(-K_1 t)\right) \tag{3}$$

With $K_4 = K_5$ and $K_1 = K_2 + K_3$ the residence time $\tau_i$, half life $T_{\frac{1}{2}i}$, decay rate $K_i$, and deposition velocity $V_i$ are related by

$$\tau_i = K_i^{-1} = \left(\frac{V_i}{h} + f\lambda\right) = 1.44\, T_{\frac{1}{2}i} \tag{4}$$

where $f$ is the fraction of time over which rain is falling, and $\lambda$ is the precipitation rate (Chamberlain, 1960; Engelman, 1968).

If the mixing height is different at time $t = 0$ and time $t = T$, the concentration computation can be adjusted with the relevant mixing height ratio given by eq. (5). The computation can also be adjusted for lateral eddy diffusion as described by eq. (6) (Scriven & Fisher, 1975). Eqs. (1)–(3) should thus be adjusted by multiplying the right hand side of the equations with the factor $\alpha\beta$ where:

$$\alpha = \frac{h(0)}{h(t)} \tag{5}$$

$$\beta = \left(1 + \frac{2\theta x}{L}\right)^{-1} \tag{6}$$

$\theta$ is the angle cross-wind spread, $x$ the travel distance to the time $t$, and $L$ the box width at time $t = 0$.

Decay rates can be determined by measurements at time $t = 0$ and time $t = T$. Assuming that $SO_4^{--}$ has the same particle distribution as trace metals, we have

$$K_4 = K_5 = T^{-1} \ln\left(\frac{r(0)}{r(T)} \alpha\beta\right) \tag{7}$$

Assuming that $K_2 \gg K_3$, we have

$$K_2 \simeq K_1 = T^{-1} \ln\left(\frac{q(0)}{q(T)} \alpha\beta\right) \tag{8}$$

$$K_3 = \frac{\{p(T)(\alpha\beta)^{-1} - p(0) \exp(-K_4 T)\}(K_1 - K_4)}{q(0)\{\exp(-K_4 T) - \exp(-K_1 T)\}} \tag{9}$$

## 10. The transport episode, February 1975

The aerosol measurements and meteorological information both from the British Isles and the Faroes during the LRT episode are now used to estimate the decay rates $K_{i=1-4}$ from eqs. (7)–(9). The air trajectories of Figs. 5 and 6 show that the air arriving at the Faroes during the episode February 25–28, 1975, did pass the British Isles one or two days earlier. Because the local wind directions at the Faroes were limited to the south-to-west clean air sector, the decrease in the gaseous and particulate concentrations can be used for estimating the decay rates for transport over water. During

this period no precipitation was reported, and the deposition was not affected by rain-out or washout. Since most particles during long range transport are usually found to be in the size interval from 0.01 to 3 $\mu$ (Fig. 6), gravitational sedimentation can be neglected. Deposition is dominated by chemical absorption and eddy impaction. These two physical effects define an eddy deposition rate and an eddy deposition velocity $K_i$ and $V_i$ used in eq. (4).

Because of the uncertainty in the average geostrophic air trajectory (Fig. 4) the transport distance $x$ in eq. (6) can vary between 800 and 1 000 km. The apparent pollution source width $L$ is estimated to vary between 200 and 400 km. $\theta$ in eq. (6) is taken to be 0.08 as suggested by Scriven & Fisher (1975). Thus we obtain $\beta = 0.6 \pm 20\%$. Anticyclonic weather conditions resulted in a subsidence inversion at the Faroes between 500 and 1 500 m, as determined by radiosonde observations which gives a typical value of 1 000 m, chosen as an average mixing height. The inversion at Van de Bilt in the Netherlands covered the whole boundary layer from zero to about 1 000 m. Reduced visibility caused by smoke was reported from several meteorological stations at the British Isles. The radiosonde observations at the British Isles showed temperature inversions about the 950 mb pressure level. Therefore the average mixing height at 55 degrees north is estimated at 500 m during the episode. This gives a mixing height correction factor $\alpha = 0.5 \pm 20\%$ (eq. 7). An average transport level is 500 m with a wind velocity at about 90% of the geostrophic 850 mb level wind.

The transport time between 55 degrees north and the Faroes, determined on the basis of the backward air trajectories, varied between 20 and 40 hours during the LRT episode. An average geostrophic transport time was 27 hours which, when corrected with respect to the height, gives a value of 30 hours.

In order to determine the decay during transport the aerosol concentration level at 55 degrees north must be determined. The British air pollution measurement station UK 2 at 55° N, 03° W gives an average during the episode of 37 micrograms per m³ air of sulphur dioxide and 13 $\mu$g/m³ of sulphate (LRTAP, monthly summary of results, NILU, Norway). The UK 2 measurement station has local sources in a distance of 1 to 10 kilometres. Measurements of sulphur dioxide from a station at Dean Moor, 54°35′ N 30°27′ W, give values of about half the UK 2 station. It should be mentioned, however, that UK 2 is analysing for total sulphur by the thorin method, and the station at Dean Moor estimates the sulphur dioxide concentration from determinations of acidity collected in the hydrogen peroxide bubbler.

Reasonable average values for the mixing layer at 55° N are here taken as 25 $\mu$g/m³ sulphur dioxide and 10 $\mu$g/m³ of sulphate.

The decay rate for sulphate is identical to that for submicron particles (Fig. 6), and it can therefore be determined from measurements of trace metal concentration e.g. lead and vanadium. Diurnal measurements of these trace metals from the British Isles are not available, however, the sources in this region can be assumed comparable to the sources in the southern part of Denmark and in the region south of Denmark (as shown by the emission survey of Eliassen & Saltbones, 1975). With comparable emission and weather conditions, results from the southern part of Denmark can be used for estimating the trace metal concentration level at 55° N over the British Isles. During a period of 140 days a measurement station at Keldsnor in Denmark—similar to that at the Faroes—provided diurnal averages of trace metals at aerosol particles. About 90% to 95% of the lead and vanadium values are below 150 and 10 nanogram per cubic metre, respectively. The higher values are observed during anticyclonic weather as during the Faroe episode in February, and the mixing height is about 500 m as estimated during the episode at the British Isles.

## 11. Eddy deposition and transformation rates

On the basis of the above description of the concentration levels February 25–28 and from the values in Table 1 we can find the eddy deposition rate $K_4$–$K_5$ from eq. (7):

$T = 30$ hours

$\alpha = 0.5$

$\beta = 0.6$

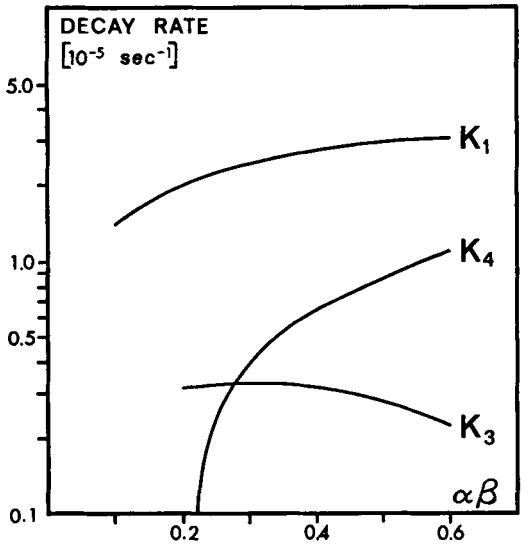


*Fig. 8.* Decay rates as function of the atmospheric dilution factor $\alpha\beta$ with measured concentrations kept constant and $\gamma = 0.3$.

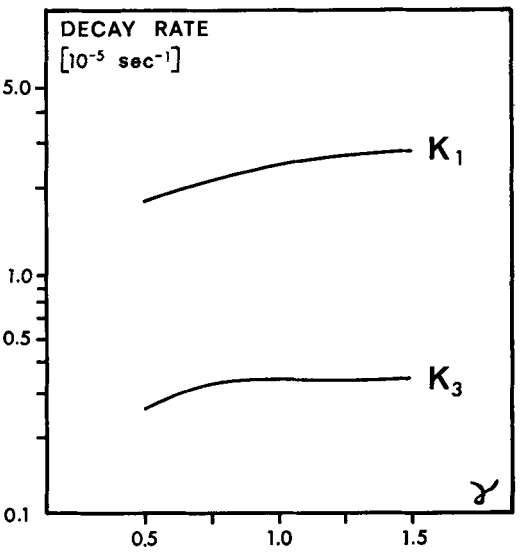


*Fig. 9.* Decay rates as function of the $SO_2$ correction factor $\gamma$ with measured concentrations kept constant and $\alpha\beta = 0.3$.

For lead and vanadium

$r(0)/r(T) = 5$

$K_4 \simeq K_5 = 0.4 \times 10^{-5}$ sec$^{-1}$

This gives a residence time of 70 hours, a half life of 48 hours and an eddy deposition velocity at 0.4 cm/sec. Using eq. (8) and Table 1, we have

$q(0)/q(T) = 46$

$K_2 \simeq K_1 = 2.4 \times 10^{-5}$ sec$^{-1}$

The average value at about $2 \times 10^{-5}$ sec$^{-1}$ gives a residence time of 12 hours, a half life of 8 hours, and an eddy deposition velocity at 2 cm/sec. The eddy deposition velocity for sulphate and sulphur dioxide is used for estimation of the transformation rate, $SO_2 \rightarrow SO_4^{--}$:

$p(0)$ = 3.3 $SO_4$–S $\mu$g/m$^3$

$p(T)$ = 1 $SO_4$–S $\mu$g/m$^3$

$q(0)$ = 12.5 $SO_2$–S $\mu$g/m$^3$

$K_1 = 2.4 \times 10^{-5}$ sec$^{-1}$

$K_4 = 0.4 \times 10^{-5}$ sec$^{-1}$

With these values eq. (9) gives $K_3 = 0.3 \times 10^{-5}$ sec$^{-1}$, which corresponds to a residence time of 90 hours and a half life of 60 hours.

The decay rates depend on the estimated atmospheric dilution factor $\alpha\beta$ as shown in Fig. 8. Using $\alpha\beta = 0.3$, $K_1$ and $K_3$ varies as shown in Fig. 9 as a function of the correction factor $\gamma$ for the $SO_2$ concentration at the Faroes, where $\gamma$ = ($SO_2$ true)/($SO_2$ measured). From this sensitivity analysis we find the error of the decay rates to be less than $\pm 50\%$, when $\alpha\beta = 0.3 \pm 20\%$ and $\gamma = 1 \pm 50\%$.

## 12. Clean Atlantic air

Table 1 reveals the average concentrations of sulphate and sulphur dioxide as 0.8 $\mu$g/m$^3$ $\pm 50\%$ and 0.2 $\mu$g/m$^3$ $\pm 50\%$, respectively, for the "cleanest possible" North Atlantic air. Our measurements are from a height of 750 m above sea level and might be taken as tropospheric average. Measurement from a ship are reported by Büchen & Georgii (1971) from the same area in the North Atlantic Ocean, giving an average value at the Faroe Islands of 0.2 $\mu$g/m$^3$ sulphur dioxide and 1.1 $\mu$g/m$^3$ sulphate. An average of 0.9 $\mu$g/m$^3$ sulphate was found over the Pacific Ocean by Dale et al. (1971). In equatorial Atlantic areas sulphur dioxide concentrations can be even less than 0.2 $\mu$g/m$^3$ (Jost, 1970). These levels are also in agreement with results from

other clean air areas reported by e.g. Coung et al. (1974) and Georgii (1970). The global sulphur cycle by Robinson & Robbins (1970), however, is based on earlier measurements of a higher "background" level, which leads to an overestimation of the natural sulphur emission to the atmosphere from the sea.

The usual assumptions concerning the natural emission of sulphur dioxide have been questioned by Beilke et al. (1974), who show that the ocean, because of its relatively high pH and good carbonate buffering action, actually is a good sink of sulphur dioxide and may not be a direct source. Rasmussen (1974) shows that ocean waters are much too oxidizing to permit the existence of hydrogen sulphide at concentrations significant to sustain transfer to the atmosphere. Thus Robinson & Robbins (1970) seriously overestimate the global free ocean sulphur emission to be 50% of the total anthropogen emission. In the future natural emission from the free ocean surfaces should be included as a considerably lesser source in the global sulphur cycle.[1] Sulphur emission as gaseous compounds from the free ocean surfaces can probably be neglected in regional dispersion budgets for West Europe, although coastal sources might be of local importance e.g. in areas with tidal effects.

## 13. Conclusion

Aerosol concentrations are measured at the Faroes during a four-day period with clean Atlantic air and a four-day period with long range transport of atmospheric pollutants over a distance of 1 000 km from the British Isles. The two periods are selected on the basis of air trajectories and a series of daily aerosol measurements. The Atlantic aerosol particle number distribution function and the concentration of trace metals are found to be in accordance with other clean air measurements.

Aerosol measurements at the British Isles are used in combination with the Faroe measurements to determine eddy deposition and transformation rates for sulphur dioxide and sulphate. We find an eddy deposition velocity for sulphur dioxide at 2 cm/sec $\pm 50\%$ and for sulphate at 0.4 cm/sec $\pm 50\%$, which corresponds to a time constant of 14 hours and 70 hours, respectively, and a half life of 10 hours and 48 hours, respectively. The transformation rate for sulphur dioxide to sulphate is equivalent to a half life at 60 hours. These estimates are average values based on several assumptions, e.g. the width of source area, the cross wind dispersion, the height of the mixing layer, average concentrations in the mixing layer, constant decay and transformation rates during the transport and so forth. The measurements were performed during stable atmospheric conditions. These results represent one of the first direct measurements of decay during distant atmospheric transport of sulphur. The decay rates are in good agreement with the investigations by Whelpdale & Shaw (1974) and Eliassen & Saltbones (1975). The deposition rates are slightly larger than those found by Shepherd (1974), Garland et al. (1974), Owens & Powell (1974), Smith & Jeffrey (1975) and Fowler (1974). The "clean Atlantic aerosol" has been measured to contain 0.2 $\mu g/m^3$ $\pm 50\%$ sulphur dioxide and 0.8 $\mu g/m^3$ $\pm 50\%$ sulphate. This is in agreement with other recent "background measurements" and suggests that previously the natural gaseous sulphur emission from oceans to the atmosphere have been overestimated in the global sulphur cycle.

Scanning electron microscopy has demonstrated distinct morphological and chemical differences between the Atlantic background aerosol particles and the aerosol particles in air which have passed the British Isles and the European continent one to three days before sampling.

[1] Our Atlantic background measurements are obtained at a cliff of 740 m height with no direct influence from coastal sources and include therefore a direct measurement of the free ocean contribution. An assumption of higher Atlantic background concentrations could only be justified in the explicit presence of coastal sources, whose global contribution is unknown at present.

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## СТЕПЕНЬ ОПАДАНИЯ И ПРЕВРАЩЕНИЯ $SO_2$ ВО ВРЕМЯ АТМОСФЕРИЧЕСКОГО ДВИЖЕНИЯ НАД АТЛАНТИЧЕСКИМ ОКЕАНОМ

Степень опадания и превращения являются основными параметрами, которые служат для определения рассеивания и опадания аэрозолей. На основе измерений в районе Фарерских и Британских островов доказано, что разница в концентрации, которая возникает во-время движения над водой на притяжении 1 000 километров, есть пропорциональна к скорости опадания во-время бурного движения, которая выносит 2 *см/сек* для $SO_2$ и 0,5 *см/сек* для $SO_4$.

Скорость превращения $SO_2$ в $SO_4$ во-время постепенного опадания при бурном движении близкий времени полураспада $SO_4$, который равен 60 часам. Чистый воздух атлантического океана содержит 0,8 микрограмма $SO_2$ на 1 *м* и 0,2 микрограмма $SO_4$ на 1 *м*, это соответствует результатам других, проведенных в последние время исследований. Такой уровень концентрации является от двух до трех раз меньше чем был ранше принятый средний уровень для тропосферы. Кроме того участие моря в натуральной выработке серы, оценивалось слишком высоко. Критерием используемых методов послужило разделение частичек аэрозола согласно их величины. Кроме того измеряются и дискутируются концентрации разных следовых элементов.