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Airborne measurements of air pollution over Denmark and the adjacent seas

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ABSTRACT

Airborne measurements of concentrations of gaseous and particulate sulphur and of Aitken nuclei were performed as part of an international project to assess the role of long-range transport of air pollution in Europe.

A description is given of the measuring equipment installed in the aircraft and some of the results are presented and discussed. A definite correlation was found between the concentrations of sulphate and sulphur dioxide; moreover, during different pollution episodes the relations between these substances were almost identical. It was also found that the concentration of Aitken nuclei correlated well with the concentration of sulphur dioxide, but that the correlation with sulphate concentration was dubious. In quite remote areas, such as over the ocean, under meteorological conditions favourable to long-range transport, it was found that pollution levels can be comparable to those encountered in large cities.

1. Introduction

The increased use of fossil fuel in Western Europe and the accompanying emission of sulphur dioxide led during the 1960's to enhanced concentrations of sulphate ions and acid in precipitation. The impact on the biosphere is considerable; thus forest growth may be threatened and a lowering of the pH in lakes and rivers can result in considerable damage to the fish fauna, as has already been seen in Scandinavia.

As materials dispersed into the atmosphere may be deposited far from their sources, it is necessary to view the problem on a continental scale. Accordingly a joint European air pollution monitoring programme was established under the auspices of the OECD to assess the role of Long Range Transport of Air Pollution (LRTAP).

It was agreed that each participating country should establish a network of stations to diurnally monitor the concentrations of sulphate and sulphur dioxide in air and the concentrations of sulphate and various salts, as well as the acidity, in precipitation. In order to obtain information on vertical pollution profiles, and to determine the pollution level over the sea, it was found desirable that the ground-level measurements be supplemented by measurements taken aboard instrumented aircraft. Each country should further provide a sulphur dioxide emission survey (Ottar & Semb, 1973; Eliassen & Saltbones 1975, Fig. 1). The programme also aimed at the construction of computer models which, on the basis of daily meteorological observations, could calculate and predict pollution levels anywhere in Western Europe. The model has been described by Eliassen & Saltbones (1975).

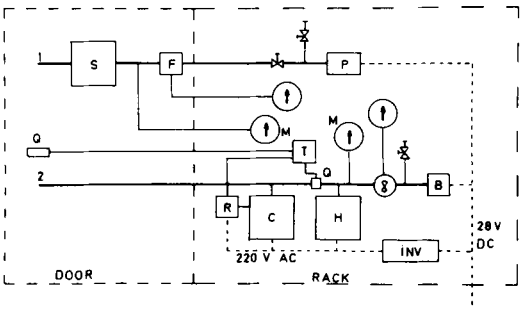

Fig. 1. Block diagram of instruments. The door: 1 and 2: Intake tubes. S: Sulphur sampler. F: Flow sensor. Q: Quartz crystal. The rack: C: Particle counter. H: Humidity meter. T: Temperature electronics. R: Recorder. M: Manometers. P: Pump. B: Blower. INV: Inverter.

Long-range transport of air pollutants is expected to be of importance when the air masses move under a horizontally extended inversion layer. Such air pollution episodes occur 6–10 times a year and last 1–3 days, in exceptional cases up to 1 week (Ottar & Semb, 1973). During meteorologically forecast episodes, the ground stations increased the activity to sampling periods of 6 hours, and a number of instrumented aircraft were alerted in the countries affected.

All the data were reported to a Central Coordinating Unit (CCU) at the Norwegian Institute for Air Research. The programme was in operation from July 1972 to March 1975.

The Danish contribution to the LRTAP project comprised six ground stations and one instrumented aircraft. The ground stations operated under the supervision of the Danish Meteorological Institute. The aircraft programme was run by the Aerosol Sciences Laboratory at Risø. The present paper describes the Danish airborne measurements system and some of the results obtained during flights over Denmark and the adjacent seas.

2. Experimental

Aircraft

The aircraft to be used in air pollution measurements must be chosen with due consideration of the capacity, availability, ease of installation of instrumentation, and economy. For the LRTAP project the aircraft should be capable of operating up to 1000 km from the base with a total payload of 500 kg and of delivering a power of 1.4 kW. These requirements call for a rather large aircraft. The availability requirement proved almost impossible to fulfil through private chartering, and therefore the aircraft was chartered from the Royal Danish Air Force, who have a number of C-47 aircraft, a military transport version of the DC-3. This aeroplane has ample capacity for the project, while its rather low cruising speed, 250 km/h, makes it ideally suited for air pollution measurements and sampling.

The availability was greatly increased by designing the equipment so that it could be installed in any of the C-47 aircraft. This was accomplished by mounting the intake tubes on a special door designed to replace an emergency door situated on the port side in front of the propeller and immediately behind the pilot's seat. The instruments with accessories were mounted compactly in a single standard 19 in. rack which could be bolted to the cabin floor. The instruments could thus be handled as one unit which facilitated the installation in the aircraft, which could be completed within 1 hour.

Instrumentation

The primary purpose of the measurements was to determine the concentrations of sulphate and sulphur dioxide. This was accomplished by collecting filter-paper samples for subsequent analysis in the laboratory. In addition, the particle concentration was measured on a direct-register instrument which was used as an indicator of the pollution level. The ambient temperature and the relative humidity of the air were also measured.

A block diagram of the equipment is shown in Fig. 1. The intake tubes, the sulphur sampler, and a thermometer were mounted on the door, while the rest of the equipment was placed in the cabin rack.

The intake tubes protruded about 10 cm in the flight direction from a dome mounted on the door; the distance from the fuselage was 15 cm, so the tubes were well outside the boundary layer surrounding the aircraft body (Megaw, 1973). The sampling orifices were designed for isokinetic sampling at an airspeed of 250 km/h. Through tapering cones the diameters were trebled to minimize losses of particles due to collision with or diffusion to the tube walls. The intake tube leading to the sulphur sampler was lined with teflon to minimize losses of sulphur dioxide due to its possible reaction with the tube material. In order to further reduce possible losses of sulphur dioxide the sampler was mounted on the inside of the door so that the tube length was kept as short as 75 cm.

The sulphur sampler collects sulphate and sulphur dioxide on two filters (Whatman 41) exposed to the air flow in series. The first filter collects particles and the second filter, impregnated with 0.5 N KOH, samples the sulphur dioxide with an efficiency greater than 90%. The sulphur contents are subsequently determined by isotope dilution analysis (Klockow et al., 1974; Flyger et al., 1976). The IDA has a lower limit of detection of 50 ng S per sample and an accuracy of 10%.

The whole sampler is contained in an airtight box with a removable lid sealed with an O-ring. The

filters are 6-cm-wide bands running from storage rolls past the exposure area to collection cassettes. The filter holders and the filter bands can be manipulated from outside the box to allow new sections of the filter bands to be exposed. The exposure area is 40 mm in diameter and provided with a teflon support grid; it is sealed with two concentric O-rings. The pressure drop over the filters is 150 mmHg. There is a small leak between the interior of the box and the interior of the filter holders. It is, however, less than 1% of the sample flow, and as the leak between the interior and the exterior of the box is less than 1‰ of the sample flow, the combined effect of these leaks is entirely negligible.

The thermometer is a small quartz crystal mounted on the door below the dome in an attempt to keep it in the shade. Adiabatic heating due to acceleration of the air in front of the thermometer causes systematic temperature errors of up to 3 °C. A correction was attempted by subtracting 1.5 °C from the readings, leading to an experimental error of the same magnitude.

The concentration of Aitken nuclei (0.005–0.2 μ) was measured with an automated version of the Nolan-Pollak condensation nuclei counter (Model Rich 100, Environment One Corporation, Schenectady, N.Y.). It has a cycle time of about 1 sec and can measure concentrations between 500 and 300.000 cm^{-3}. A signal proportional to the particle concentration is recorded continuously. The pneumatic control system of the instrument had to be modified slightly to work properly under reduced ambient pressure.

The counter is a volume-controlled instrument: the readings are obtained after an adiabatic expansion of the sampled, humidified air from one fixed volume to a larger fixed volume. Therefore measurements on this counter do not, in principle, depend on the ambient pressure, i.e. the flight altitude, except that the concentrations registered refer to ambient volumes (Nolan & Scott, 1964). However, at ambient pressures below 800 mb corresponding to altitudes above 2000 m, the results are considered to be indicative only.

Operational procedures

The preparations for and the execution of alerted flights generally followed the same predetermined pattern: the CCU issued a first warning 48 hours prior to take-off and it was followed by an affirmative warning or a cancellation 24 hours later. In the meantime a survey of possible flight routes was carried out in cooperation with the Meteorological Institute and the instruments were checked. The flight was normally divided into two phases. The first phase, performed en route to the starting position, consisted of measurements of pollution levels and temperature during a stepwise increase in altitude in an attempt to map out the vertical profiles. The second phase consisted of the measurements proper. A flight normally lasted between 3 and 6 hours.

3. Results

The results obtained on three flights in February 1975 are presented below. The flights were numbered 7501, 7502 and 7503 and were executed on February 6, 21 and 22. The two latter flights were performed during the same episode and will to some extent be treated as one flight. For each of these days isobaric trajectories have been calculated using two-dimensional geostrophic winds. The trajectories go 48 hours back in time in steps of 3 hours. No account was taken of the topography or of the effect of surface friction.

Flight 7501

The meteorological situation was characterized by a high centred over the North Sea; this gave rise to a predominantly anticyclonic circulation. Details of the circulation can be seen in Fig. 2, where the trajectories in the 1000 mb and the 850 mb surfaces are shown. The air arriving in the southwestern corner of the North Sea is seen to have passed over areas with large SO_2-emissions within the preceding 12 hours. This is not the case for the 1000 mb trajectories terminating over Denmark in the morning and the afternoon of February 6. The corresponding 850 mb trajectories have passed over the northern part of the North Sea and clearly show the anticyclonic circulation. Measurements on an E–W cross section of the North Sea could therefore be expected to reflect the different history of the air masses. The flight route was accordingly chosen as shown in Fig. 3; the flight altitudes were 150–300 m above sea level but, due to the cloud, 600 m at the beginning (sample 1) and end (samples 9 and 10) of the flight.

The sulphur concentrations are listed in Table 1 together with values supplied by CCU. These are

ground level concentrations and values calculated from the dispersion model. The ground level values have been estimated for the sampling positions from isopleth-diagrams for Europe, based on all the 24-hour samples taken at the LRTAP stations. For flight 7501 the distances to the nearest stations are 100 km or more.

The concentrations of Aitken nuclei, sulphur dioxide and sulphate, denoted henceforth Z, SO_2 and SO_4, respectively, are plotted chronologically in Fig. 4 together with the flight altitudes. The concentrations were, as expected, much higher over the central and southwestern part of the North Sea than over the eastern part of the North Sea and Denmark. This is particularly true for SO_2 which reached levels that are of the same order of magnitude as those found in large cities. The concentration of sulphate remained constant at a high level during the central part of the flight; in samples 1, 2 and 10 sulphate concentrations were below the detection limit. The temperaure varied between 5 °C and 11 °C with a tendency for the higher values to occur over midocean and at the higher altitudes. The relative humidity was generally be-

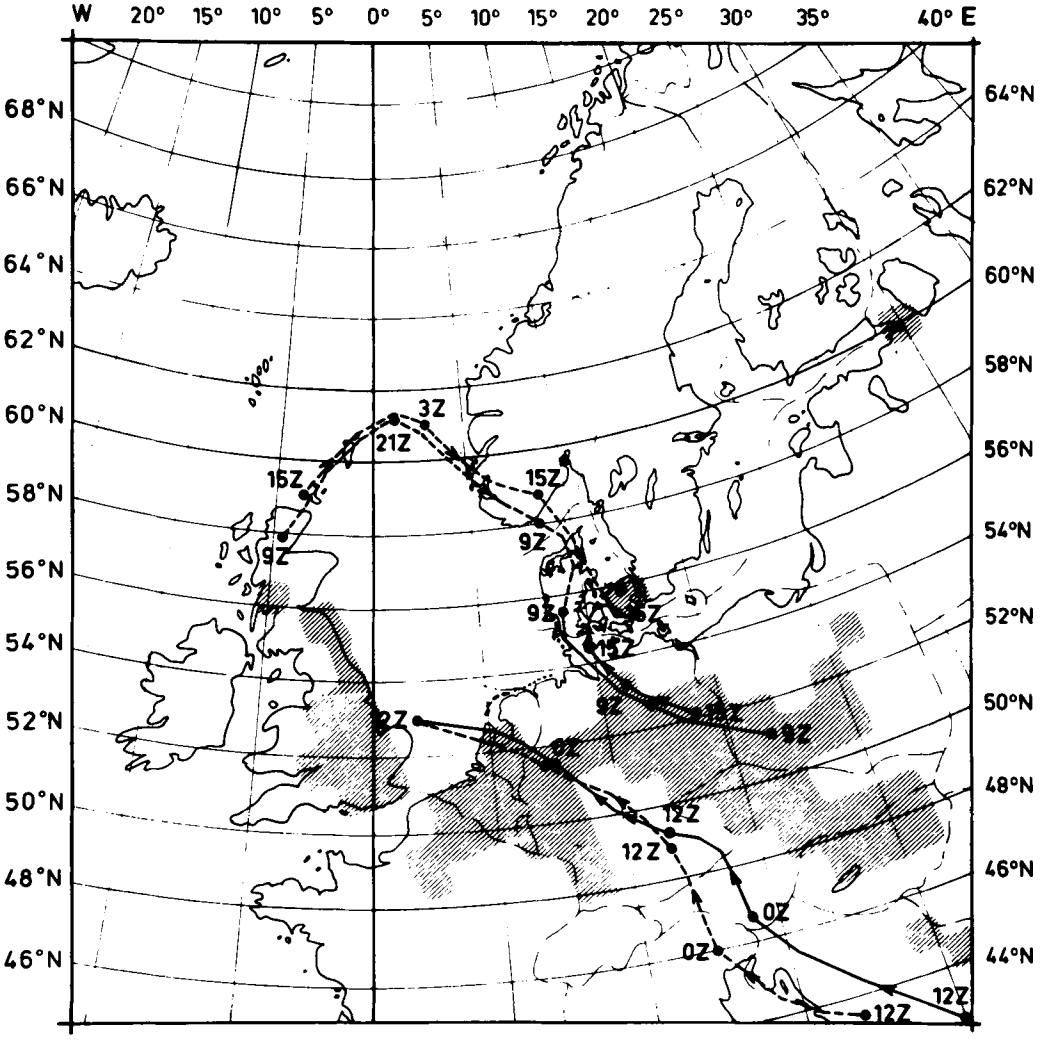

Fig. 2. Isobaric trajectories for February 6, 1975, flight7501. Full lines: 1000 mb. Dashed lines: 850 mb. *Z* refers to GMT. In the hatched areas SO_2-emission exceeds 10 t/km^2-year (Eliassen & Saltbones, 1975, Fig. 1).

tween 50% and 60%, but a value as low as 20% occurred during sample 5.

A geometric mean particle concentration, Z_g, has been calculated for each sampling period, using readings every second minute. A statistical analysis shows that positive correlations, significant at the 95% level, exist between SO_4 concentrations on the one hand and SO_2 concentrations and Z_g on the other hand; SO_2 concentrations are also positively correlated with Z_g.

The orthogonal regression line between the concentrations of SO_4 and SO_2, expressed in $\mu g/kg$, is:

$SO_4 = 0.13 \cdot SO_2 + 1.66$

and the correlation coefficient is $r = 0.81$.

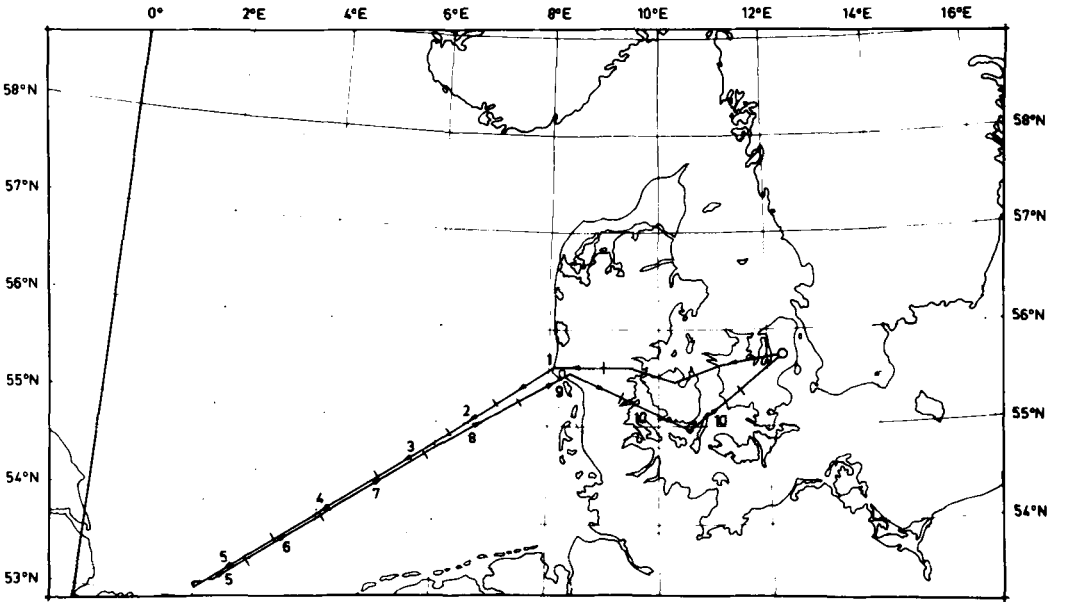

Fig. 3. Route for flight 7501 indicating the approximate location of the samplings.

Table 1. *Concentrations of particulate and gaseous sulphur measured in the aircraft, on the ground, and calculated from the model. February 6, 1975; flight 7501*

	SO_4 ($\mu g/kg$)			SO_2 ($\mu g/kg$)		
Sample	Aircraft	Ground	Model	Aircraft	Ground	Model
1	–	2	2	3	14	4
2	–	3	2	3	15	6
3	3	8	1	8	15	20
4	9	12	12	25	23	36
5	8	15	12	60	46	43
6	8	12	12	42	39	42
7	3	6	11	20	15	36
8	2	2	3	3	15	3
9	0	2	1	3	14	5
10	–	5	2	2	23	9

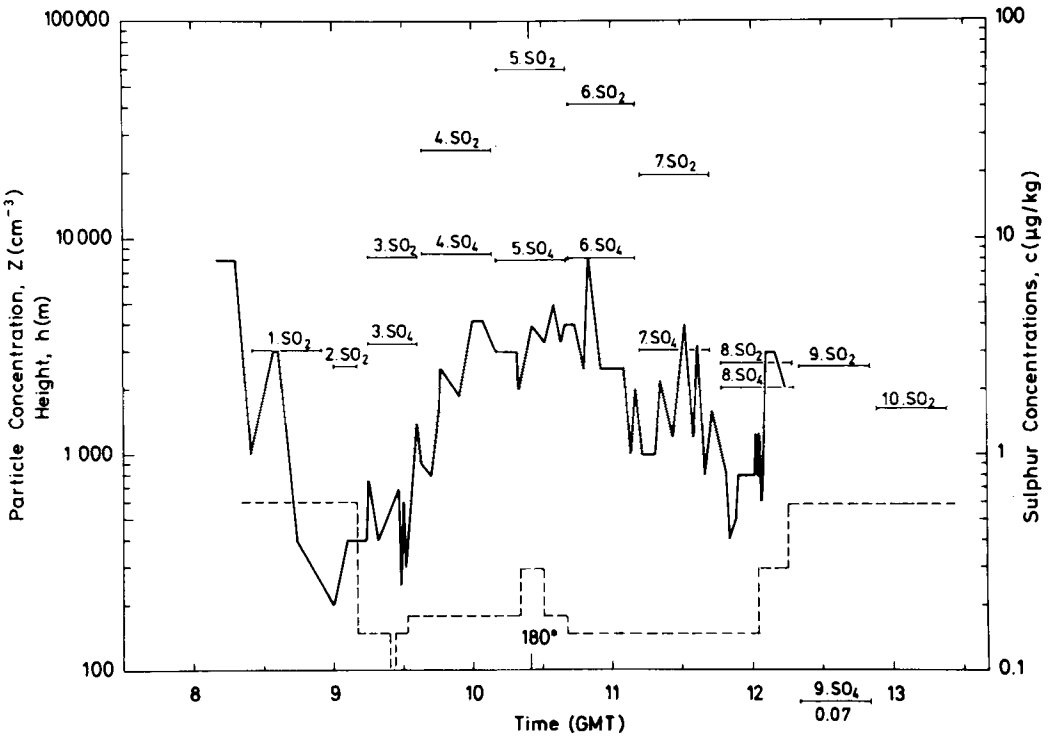

Fig. 4. Chronological plot of the results obtained during flight 7501. Left axis: Aitken nuclei concentrations Z (full line) and heights in m (dashed line). Right axis: Concentrations of sulphate (SO_4) and sulphur dioxide (SO_2). The number and duration of the samples are indicated.

Flights 7502 and 7503

On February 21 a high was located southeast of Denmark; during the night and the following day it moved slowly further southeast. This gave rise to an anticyclonic circulation over the northern part of Central Europe. Isobaric trajectories at 1000 mb are shown for the 2 days in Fig. 5. The air arriving over Denmark during this episode is seen to have passed over areas with high SO_2-emissions. The flight routes were chosen perpendicular to the wind, so that flight 7502 crossed the country from northwest to southwest at altitudes between 300 m and 450 m, whereas flight 7503 followed the 55 N latitude from 15 E to 7.5 E at altitudes between 150 m and 450 m.

In Fig. 6 are shown the profiles of temperature and particle concentration measured at the beginning of flight 7503. A temperature inversion seems to have existed below 600 m, particularly pronounced below 300 m, where the particle concentration was quite high and where a sudden jump indicates the existence of a pollution layer. At 1200 m the simultaneous jumps in temperature and Aitken nuclei concentration may also mark the beginning of another layer.

The results from flight 7503 are plotted chronologically in Fig. 7. The concentrations of sulphate and sulphur dioxide were quite high and did not exhibit the large systematic variations encountered in flight 7501; this is what could be expected considering the meteorological situation and the sampling positions. The only exception to this was the last sample taken at a height of 1800 m: the SO_2 concentration was small, as could be expected, whereas the SO_4 concentration was surprisingly large.

As noted in connection with the profiles, the pollution seemed to be stratified. This is to some extent confirmed by the large sudden variation in particle concentrations which occurred even at constant altitudes; values as high as 30 000–40 000 cm^{-3} were encountered during these excursions. If the aircraft had been skimming the top or the bottom of a pollution layer the particle concentration would

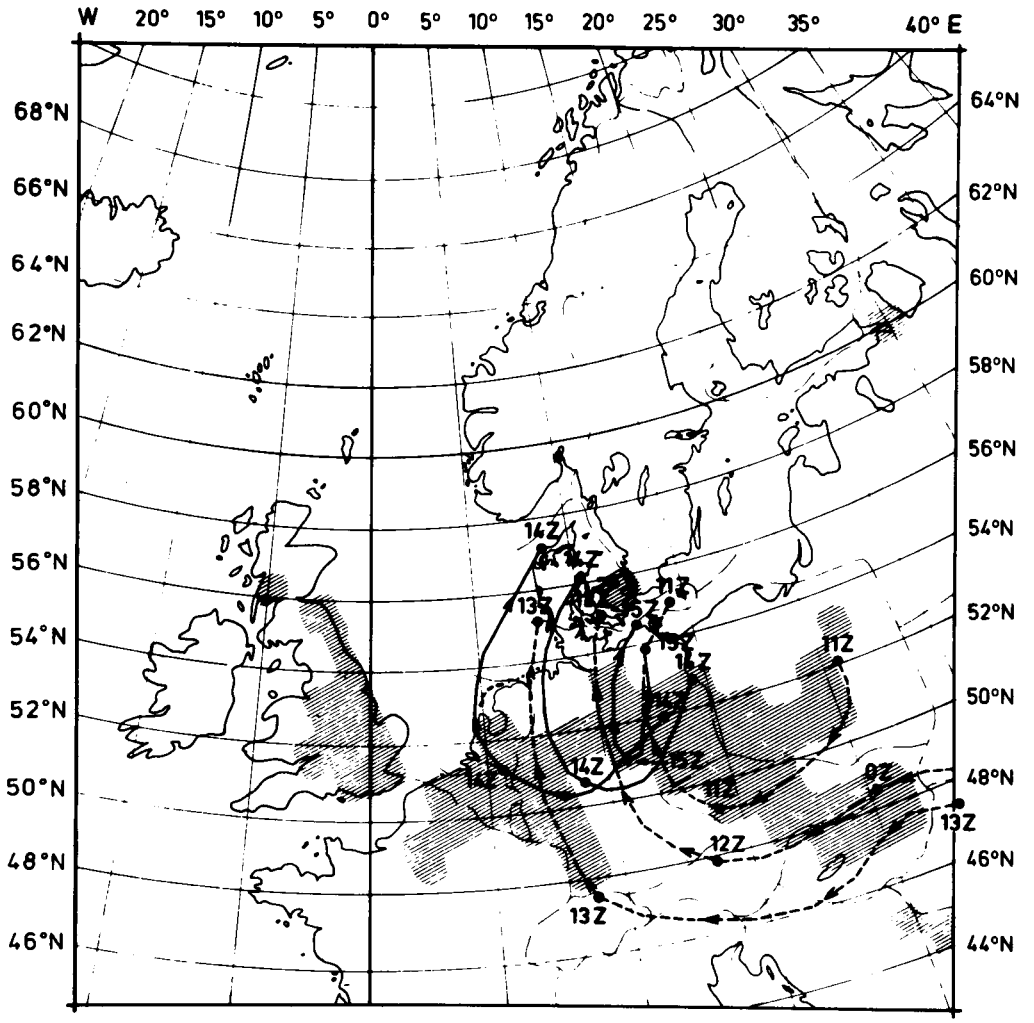

Fig. 5. Isobaric trajectories at 1000 mb. Full lines: February 21, 1975, flight 7502. Dashed lines: February 22, 1975, flight 7503.

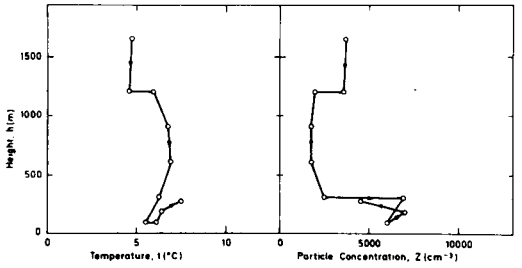

Fig. 6. Profiles of temperature and particle concentration measured on flight 7503.

certainly exhibit large variations. In an attempt to stay in the layer the height was changed several times, as can be seen in Fig. 7.

The sulphur concentrations from the two flights are listed in Table 2 together with ground level results and values calculated from the model. The results from the three samples taken on flight 7502 do not differ markedly from the results from flight 7503; the relative humidity was, however, quite low and varied between 15% and 30%.

A correlation analysis similar to that described for flight 7501 reveals that for neither of the two

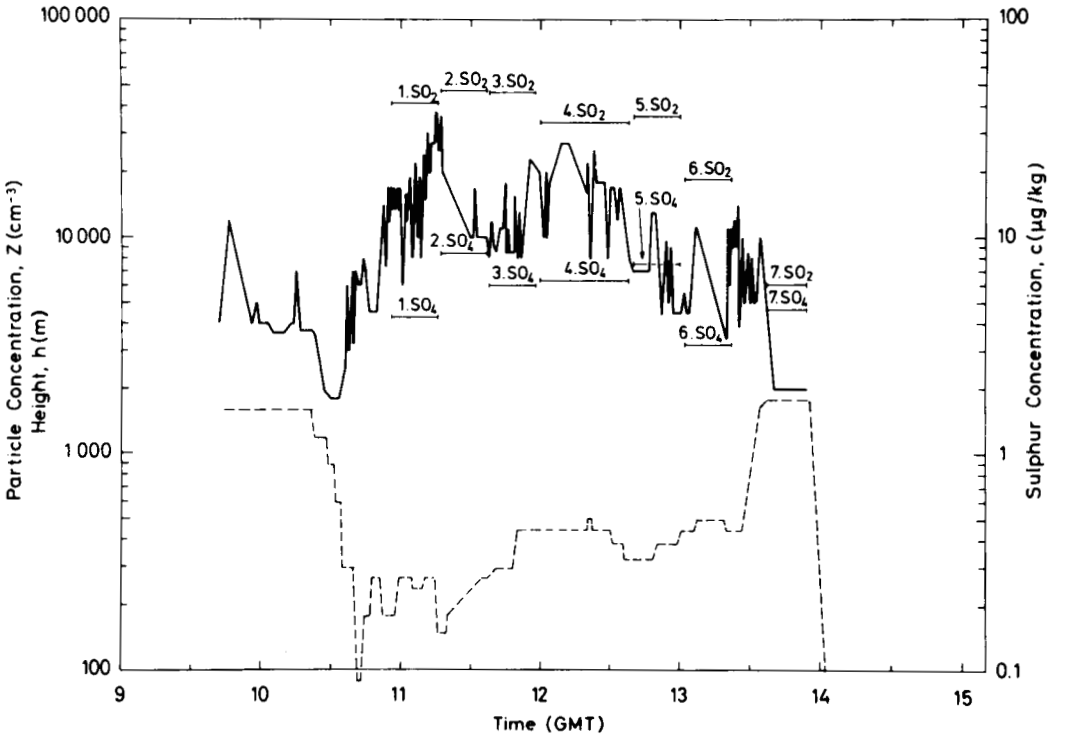

Fig. 7. Chronological plot of the results obtained during flight 7503. Legend: Same as Fig. 4.

Table 2. *Concentrations of particulate and gaseous sulphur measured in the aircraft, on the ground, and calculated from the model. February 21, 1975, flight 7502, and February 22, 1975, flight 7503*

	SO_4 (μg/kg)			SO_2 (μg/kg)		
Sample	Aircraft	Ground	Model	Aircraft	Ground	Model
1	2	11	5	11	19	25
2	3	10	4	30	22	31
3	0	7	2	14	9	31
1	4	10	4	41	31	39
2	9	12	4	47	33	27
3	6	15	3	46	36	29
4	6	15	4	34	37	18
5	8	14	3	36	46	20
6	3	15	3	18	37	20
7	5	16	3	6	27	16

flights taken separately were the concentrations of the three pollutants significantly correlated above 95% confidence, with the exception of SO_2 and Z_g for flight 7503.

As the two flights were performed on consecutive days during the same episode, an attempt has been made to treat them as one flight. For the combined results, significant positive correlations

were found between SO_2 on one hand and SO_4 and Z_g on the other. The orthogonal regression line between the concentrations of SO_4 and SO_2, expressed in $\mu g/kg$, is:

$$SO_4 = 0.13\ SO_2 + 0.94,$$

and the correlation coefficient is $r = 0.67$ for $n = 10$ data points, which is significant at 95% confidence. For 7502 $(r, n) = (0.65, 3)$ and for 7503 $(r, n) = (0.54, 7)$ neither of which is significant.

No significant correlation was found between particulate sulphur and the Aitken nuclei concentrations.

4. Discussion

The episode flights were part of a large international investigation and were executed for a very specific purpose. When the results are not considered in this context but isolated as in this paper, they allow no general conclusions. However, a few points of interest should be noted.

Flight 7501 was carried out in a rather ideal situation which allowed measurements inside and outside a plume coming from the European continent. This is reflected in the systematic variations in pollution levels, notably the SO_2-concentration, which in the centre of the plume is 30 times higher than outside the plume (Table 1). This clearly demonstrates the importance of long-range transport of pollution: the measurements were taken over the open sea with no interference from local sources. Actually this flight proved to be one of the more successful ones within the LRTAP project, in that it confirmed the model calculations in an area from which no measurements were available prior to the episode (Nordø, 1975). Unfortunately no other LRTAP flights were carried out on this particular day. The correlation coefficients for the series of sulphur concentrations listed in Tables 1 and 2, obtained in the air (A), estimated ground concentrations (G), and as calculated model values (M) are given in Table 3. For flight 7501 the three series of data correlate well for both particulate and gaseous sulphur, with the exception of ground and model results for SO_2.

For the combined results of flights 7502 and 7503, and also for the results of 7503 alone, the data cannot be said to correlate. In contrast to flight 7501 these two flights did not include measurements outside the plume and furthermore the pollution seemed to be stratified. Two other LTRAP flights, N34 and N35, were carried out over the North Sea by Norway simultaneously with flights 7502 and 7503, respectively. The airborne, ground and model results for N34 and N35 do not correlate either. The data are, however, too few and cover too short a time to reflect the general, large-scale validity of the numerical model.

The non-significant correlation coefficients, which even come out negative in some cases, reveal the gross discrepancies, which are often found between airborne and surface measurements. Stratification of pollution, which has been found both in this investigation, in other flights within the LRTAP project (Gotaas, 1975b) and in flights over Greenland (Flyger et al., 1976), makes it difficult, if not impossible, to infer the pollution levels aloft from surface measurements alone. This is further complicated by the rather long sampling times used at ground stations. In order to set up a reliable dispersion model it is necessary to supplement the long-term surface measurements with short-term measurements which contain some sort of vertical profile. It is also necessary to get results from the large sea areas where no regular measurements are taken at all. Hence the importance of airborne measurements.

The concentrations of sulphate and sulphur dioxide were linearly related during these two episodes and it is noteworthy that the residual constants are

Table 3. *Correlation coefficients for the data of Tables 1 and 2*

	SO_4			SO_2		
Flight no.	*A–G*	*G–M*	*M–A*	*A–G*	*G–M*	*M–A*
7501	0.93	0.81	0.84	0.92	0.59	0.89
7502 + 7503	0.57	–0.02	0.14	0.56	–0.38	0.17

rather small compared to typical SO_4 values and that a regression coefficient of 0.13 appears in both expressions. Thus the two regression lines are almost identical. Another striking feature is that a regression coefficient of 0.13 was also found in airborne measurements over the oceans around Greenland (Flyger et al., 1976). The correlation between SO_2 and SO_4 can be explained by assuming that the process of converting sulphur dioxide to sulphate has, both in quite clean air and in heavily polluted air, reached approximately the same stage of progress. The absence of a correlation would then mean that the assumed process was encountered at widely differing stages of progress, or that recent influxes of either SO_2 or SO_4 had occurred along the trajectories. With the few data presented here it cannot be excluded, however, that the correlations are fortuitous. In this connection it should be noted that the concentrations of sulphate and sulphur dioxide measured on the two flights N34 and N35, carried out simultaneously with 7502 and 7503, do not correlate well.

The Aitken nuclei concentrations are apparently much better correlated with the concentrations of gaseous sulphur than with the concentrations of particulate sulphur. It should be recalled, however, that the sulphate concentration measures the mass of the sulphur particles and this is dominated by the aged particles; the mass median probably lies around 0.5 μ (Meszaros, 1973; Georgii et al., 1971). The Aitken nuclei concentration refers, on the other hand, to the small particles and it is a number concentration that is likely to be dominated by the very small particles, say less than 0.05 μ as has been found in a background area such as Greenland (Flyger et al., 1976). At the concentrations measured, such small particles cannot survive for more than a few hours before they coagulate into larger ones (Zebel, 1966). They are therefore thought to originate locally as a result of gas phase reactions. Thus the significant correlation between the concentrations of sulphur dioxide and Aitken nuclei suggests that a large fraction of the latter originates from the oxidation process of sulphur dioxide.

The results presented above and those obtained on flights in other countries (Smith & Jeffrey, 1975; Gotaas, 1975a, b), show clearly that long-range transport of air pollution is a factor of importance. In remote areas, e.g. over the sea more than 100 km from the coast, pollution levels are normally very low and often comparable to true background levels in very remote locations, such as Greenland (Flyger et al., 1976). In certain meteorological situations long-range transport of pollution may, however, give rise to concentrations in these areas that are comparable to those encountered in large cities.

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САМОЛЕТНЫЕ ИЗМЕРЕНИЯ ЗАГРЯЗНЕНИЙ ВОЗДУХА НАД ДАНИЕЙ И ОКРУЖАЮЩИМИ МОРЯМИ

В качестве раздела международного проекта исследований роли крупномасштабного переноса загрязнений воздуха в Европе были выполнены измерения концентраций серы в газообразной итвердой фазе и ядер Айткена. Даётся описание оборудования для измерений, установленного на самолёте. Представляются и обсуждаются некоторые результаты измерений. Найдена определённая корреляция между концентрациями сульфатов и двуокиси серы; более того, в течение различных событий загрязнения соотношения между этими субстанциями были почти идентичными. Также было обнаружено, что концентрация ядер Айткена хорошо коррелировала с концен трацией двуокиси серы, но корреляция с концентрацией сульфатов была сомнительной. Было найдено, что в довольно отдалённых областях, таких, как над океаном, при метеорологических условиях, благоприятствующих крупномасщтабному переносу, уровень загрязнений может быть сравним с тем, который встречается над большими городами.