

A STUDY OF THE RATIO BETWEEN DOMESTIC AND FOREIGN CONTRIBUTIONS TO HIGH SULFUR CONCENTRATIONS IN THE AIR, MEASURED IN FINLAND MARCH 25–26, 1973

by

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A b s t r a c t

The calculations based on dispersion modeling, were performed in order to obtain the most probable and maximum possible values for the domestic contributions. The results showed that only a small part of the sulfur (SO_2 and SO_4) concentrations measured probably originated from domestic sources. Even the estimated maximum possible domestic contributions were not more than about 50 % of the measured values. The main part of the sulfur concentrations observed must therefore have been of foreign origin. According to the calculations, this foreign sulfur was causing pollution concentrations of 12–42 $\mu\text{g SO}_2/\text{m}^3$ and 7–25 $\mu\text{g SO}_4/\text{m}^3$ in the inner areas of Finland. Compared with long-term averages and global background concentrations these values must be considered very high.

1. *Introduction*

In a previous investigation (NORDLUND [11]) numerically calculated concentrations of sulfur dioxide (SO_2) over western Europe for the time period March 23–26, 1973 were presented. According to these calculations considerable amounts of sulfur were transported from the industrial areas of western and central Europe to Scandinavia and Finland. For the day March 25 the model gave SO_2 concentrations for Finland of 30 $\mu\text{g}/\text{m}^3$ in the south-west and about 20 $\mu\text{g}/\text{m}^3$ in central Finland. The concentrations measured on the same day varied at the different stations between 14 and 49 $\mu\text{g}/\text{m}^3$ for SO_2 and 7–27 $\mu\text{g}/\text{m}^3$ for sulfate (SO_4). The concentrations calculated and measured agreed relatively well and this indicated that long distance transport of sulfur, capable of causing considerable concentrations

may, indeed, have occurred. Model calculations do, however, involve several sources of error and the question may still be raised whether such significant transport of sulfur really occurred. This question is further justified because four of the five measurement points in Finland were located inland and were influenced by domestic sources.

An investigation of the influence on sulfur concentrations in Jokioinen from local and domestic sources, made some years ago (BREMER [2]), indicated that the measured values correlated relatively highly with domestic sources. However, the correlation of 0.58 for a one month period observed in Bremers investigation is not surprising, since in Jokioinen there are several big sources which with certain wind directions give significant concentrations at the measurement point.

The aim of the present investigation was to determine the most probable and maximum possible contributions from domestic sources to the actual sulfur concentrations and to estimate the corresponding foreign contributions.

2. Measured sulfur concentrations and weather conditions during the period March 23–26, 1973

The 24h-concentrations, 08–08 local time (LCT), of SO₂ and SO₄ measured at the five regional air pollution measurement stations in Finland («OECD-stations»; OECD [12]) during the period March 23–26, 1973 are given in Table 1. The same table also gives the mean values for March 1973–1976.

Table 1. 24h concentrations (08–08 LCT) of SO₂ and SO₄ measured during the period March 23–26, 1973. In the last column the corresponding mean values for March 1973–1976, are given.

Station (geographical co-ordinates)	Concentrations measured ($\mu\text{g}/\text{m}^3$) on:								Mean March 1973–1976 SO ₂ SO ₄	
	March 23		March 24		March 25		March 26			
	SO ₂	SO ₄	SO ₂	SO ₄	SO ₂	SO ₄	SO ₂	SO ₄		
Jomala (60°11'N, 19°59'E)	0	7	6	9	27	21	3	5	9	3.3
Jokioinen (60°49'N, 23°30'E)	15	1	5	0	14	7	13	2	11	3.2
Puumala (61°34'N, 28°04'E)	10	0	3	3	30	10	13	4	8	2.4
Ähtäri (62°33'N, 24°13'E)	0	1	0	4	49	27	6	10	8	2.4
Sodankylä (67°22', 26°30'E)	0	1	0	3	6	10	3	1	3	1.4

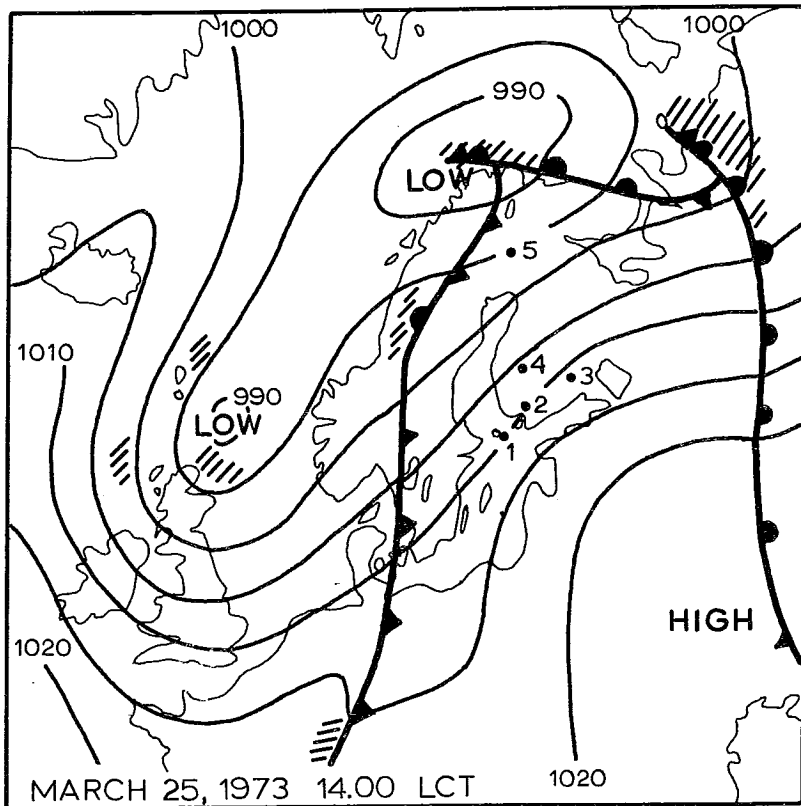


Figure 1. The synoptic surface weather map for north-western Europe for March 25, 1973 12 GMT. The isobars are drawn at 5 mb intervals. The map also shows the location of the five Finnish regional air pollution measurement stations (OECD-stations; OECD [12]), which are indicated by the numbers 1 to 5 (1 = Jomala, 2 = Jokioinen, 3 = Puumala, 4 = Ähtäri and 5 = Sodankylä).

The locations of the measurement stations are shown in Figure 1.

At the beginning of the period the weather situation was characterized by a strong high extending from the British Isles to the Black Sea. In Scandinavia and Finland there were westerly winds bringing clean air from the Atlantic. On the 24th a low moved from mid-Atlantic to Scotland and a south-westerly air current developed over western Europe and Fennoscandia. In Finland the south-westerly winds continued until the morning of March 26.

Considering regional air pollution transport, it was significant that the air over central and western Europe, which for many days had been fairly stationary, began on the 24th to move to Scandinavia and Finland:

During the period studied there was no precipitation in Finland apart from light, temporary snow in the most northern parts of the country. Figure 1 shows the synoptic weather map for March 25, 1973, 12 GMT (14 LCT in Finland).

3. *The calculation method*

3.1 A general description

To determine which domestic sources could have contributed to the sulfur concentrations measured at Jokioinen, Ähtäri and Puumala, backward air trajectories were calculated from these stations every 3 hours (Jomala was considered to be uninfluenced by domestic sources and the concentrations at Sodankylä were very small). Each trajectory was correspondingly assumed to be valid for a 3-hour period. The domestic contribution to the sulfur concentration at the measurement point, defined as the location of the sampler, was then calculated according to sources along or so close to the trajectory that they could have caused a concentration greater than $0.01 \mu\text{g}/\text{m}^3$ at the measurement point. (Concerning the distinction between small and big sources see Chapter 4.)

From each particular source the plume axis was assumed to be parallel to the actual trajectory. The concentration contribution at the measurement point was determined by multiplying the emission of the source by a relative concentration value obtained from the estimated horizontal and vertical concentration distribution in the plume. A Gaussian concentration distribution was assumed in the horizontal. For sources nearer than 10 km from the measurement point, the Gaussian approximation was also used in the vertical direction. For more distant sources the vertical concentration distribution was calculated separately by using a gradient-transfer method.

The concentration contributions from the single sources were added in order to obtain the concentration value at the measurement point for one trajectory, representing a 3-hour period. The mean concentration for the 24-hour period was obtained as an arithmetic mean of the values of the 3-hour periods.

3.2 Specifications

3.2.1 The trajectory calculation

The air trajectories were calculated backwards from the measurement points at 08, 11, 14, 17, 20, 23, 02, 05 and 08 LCT corresponding to the synoptic observation times. For each trajectory calculation a »main transport level» was determined. This main transport level was assumed to be half the height of the

mixing layer, which in turn was determined as the height to the first upper inversion in the routine radiosonde observations in Jokioinen (used for Jokioinen) and in Luonetjärvi near the town of Jyväskylä (used for Puumala and Ähtäri). If there was a ground level inversion, a height of 200 m was used as the main transport level, corresponding to the estimated effective release height of major distant sources. The wind values for the main transport level used, were calculated by applying a correction for wind shear (corresponding to the shear observed in the soundings at Jokioinen and Luonetjärvi) to the anemometer-level wind value analysed from the synoptic observations. Since the sampler was only $1\frac{1}{2}$ m above the ground, the trajectory for the air coming to it was assumed to follow the wind at a lower level than the main transport level for the last kilometers. Therefore, the trajectories were calculated by using the wind value at the anemometer height for distances less than 2 km from the sampler and by using only half of the shear correction between 2 and 20 km.

The wind values for the anemometer height were calculated by interpolation from the three best located synoptic wind observations surrounding the endpoint of the trajectory at the actual time-step. The time-step was 10 minutes and consequently after each 10 minutes the transport wind was analysed again.

The displacement (backwards in time) of the trajectory endpoint during one time-step was calculated by:

$$x^{n+1} = x^n - u^n \Delta t; \quad y^{n+1} = y^n - v^n \Delta t, \quad (1)$$

where the index n refers to the ordinal number of the time-step, x and y to the coordinates of the trajectory endpoint, and u^n and v^n to the wind components at the point (x^n, y^n) . Linear time interpolation was used between each 3-hour synoptic wind observation. The trajectories were calculated backwards in time until they had left the Finnish mainland. Sources in the Åland Islands were therefore not considered as domestic. The influence of these sources would in any case have been very small.

3.2.2 The horizontal and vertical concentration distributions

Horizontal diffusion

Horizontally, the common Gaussian plume approximation was used, and the concentration distribution from one particular source described by*:

$$\chi_h(x, y) = \frac{1}{\sqrt{2\pi} \sigma_y(x) \bar{u}} \exp\left(-\frac{y^2}{2\sigma_y(x)^2}\right), \quad (2)$$

* By multiplying (2) and (6) the common Gaussian plume equation is obtained. This equation is here only split up in a horizontal, χ_h , and a vertical, χ_z , concentration distribution.

where x is the distance from the source, measured along the trajectory, and y the distance from the plume axis, which was assumed to follow the trajectory. It is assumed here, that the plume axis from a source located at some distance from the trajectory, would have missed the measurement point by the same distance as the trajectory the source. It is perhaps worthwhile pointing out that there are no errors due to time displacement, since the trajectories from the measurement points were calculated backwards in time. The mean wind velocity \bar{u} in (2) was estimated for the emission height using the power expression:

$$\bar{u}_H = \bar{u}_{an} \left(\frac{z_H}{z_{an}} \right)^m \quad (3)$$

where the index H refers to the emission height and the index an to the height of the anemometer. The value 0.3 was used for m in neutral stratification and the value 0.4 in stable. These values for m , originally assigned to FROST [6], have commonly been used in air pollution investigations. The \bar{u}_{an} -values were taken from the wind observations at the synoptic stations closest to the measurement points. The values for $\sigma_y(x)$ in (2) were primally taken from the »Turner Workbook» (TURNER [20]) curves up to distances of 100 km. For longer distances the following expression was used:

$$\sigma_y(x) = A\sqrt{x}, \quad (4)$$

where the factor A was determined by matching $\sigma_y(x)$ with the Workbook σ_y -value at 100 km.

Since the trajectories were calculated at 3-hour intervals, the Workbook $\sigma_y(x)$ values, valid for a sampling time of 10 minutes (TURNER [20]) had to be corrected for this longer sampling time. A recent paper (PASQUILL [14]) notes that the Workbook σ_y -curves are most suitable for a sampling time of 3 minutes only. However, throughout the world the curves have been used for sampling times of up to one hour and therefore the validity value given by Turner was used in order to avoid overestimated $\sigma_y(x)$ values. Contours (Figure 2) drawn by F.B. Smith were used for the sampling time correction. These contours are based on large sets of data from Porton, O'Neill and Brookhaven (F.B. SMITH, private communication).

In cases where there was a significant wind shear in the vertical, the influence of this wind shear on $\sigma_y(x)$ was estimated according to the methods presented by SAFFMAN [17] and SMITH [18]. The final $\sigma_y(x)$ values were calculated from the contribution due to turbulence and vertical wind shear using:

$$\sigma_y(x) = \sqrt{(\sigma_y(x))_T^2 + (\sigma_y(x))_S^2} \quad (5)$$

where the indices T and S stand for turbulence and shear, respectively.

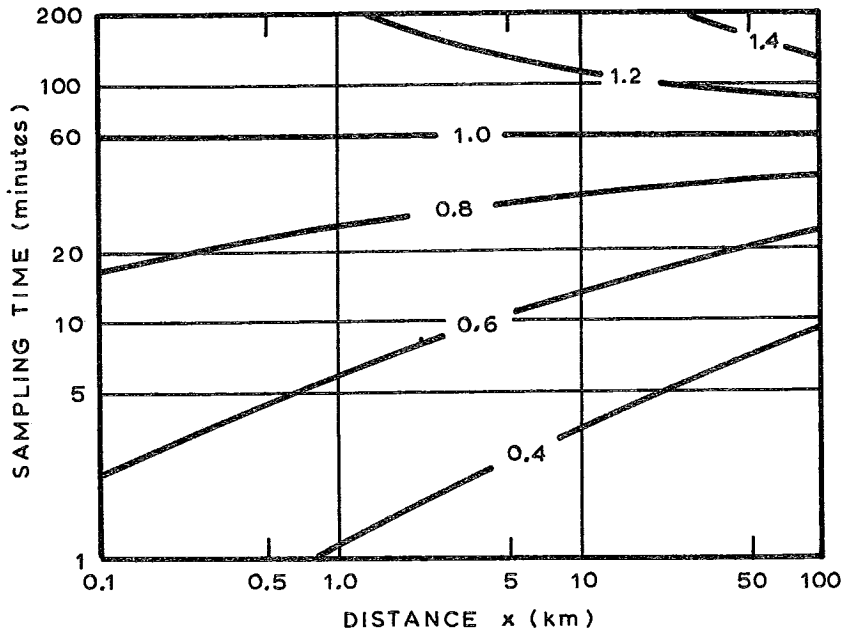


Figure 2. Contours for correction of $\sigma_y(x)$ for sampling time (after F.B. SMITH).

Vertical diffusion

For sources closer than 10 km to the measurement point in the vertical direction, a Gaussian concentration distribution was assumed, according to the chimney formula:

$$\chi_z(x, z) = \frac{1}{\sqrt{2\pi} \sigma_z(x)} \left[\exp\left(-\frac{(z+H)^2}{2\sigma_z(x)^2}\right) + \exp\left(-\frac{(z-H)^2}{2\sigma_z(x)^2}\right) \right], \quad (6)$$

where H is the effective chimney height.

To determine $\sigma_z(x)$, curves based on the Ågesta measurements in Sweden (HAEGGBLOM *et al.* [9], HÖGSTRÖM [10]) were used. The terrain at Ågesta corresponds to the type of terrain in Finland. (Since the Ågesta curves are drawn only for distances up to 10 km, they were not used in determining the horizontal $\sigma_y(x)$ values). Only the neutral and stable stability classes were used. According to radiosonde observations in Jokioinen and Luonetjärvi and to the daily temperature variation in the surface observations, the stratification was estimated to be neutral from 8.00–20.00 LCT and stable during the night.

When the transport distance is greater than about 10 km, the dry deposition starts to have a significant influence on the concentrations, and the Gaussian

approximation is no longer suitable for describing the vertical concentration distribution (e.g. PRAHM *et al.* [15]). Instead of (6) a simple gradient-transfer method for determining the vertical concentration distribution within the mixing layer was therefore used. The method was based on numerical time integration (over the transport time) of the vertical diffusion equation:

$$\frac{\partial \chi_z(z, t)}{\partial t} = \frac{\partial}{\partial z} \left(K(z) \frac{\partial \chi_z(z, t)}{\partial z} \right), \quad (7)$$

where $K(z)$ is the vertical eddy diffusion coefficient. The values for $K(z)$ were taken from standard $K(z)$ profiles for different stability types drawn by F.B. Smith (PASQUILL [13], page 345).

The solution to (7) is sensitive to numerical errors when using finite difference methods. Here Crank-Nicholson's (RICHTMYER [16]) implicit difference scheme is applied. This scheme consumes somewhat more computer time than simpler explicit schemes, but gives a fairly accurate solution.

The dry deposition on the ground was taken into account as a boundary condition by using, at ground level (in the calculations at a height of 1 m above the real ground), a sink according to the formula:

$$\chi_{z=0}^{n+1} = \chi_{z=0}^n \left(1 - \frac{\nu_d \Delta t}{\Delta z} \right), \quad (8)$$

where n refers to the ordinal number of the time-step, Δt the time-step (10 s in the profile calculations) and Δz the vertical grid point distance (used value 10 m). ν_d is the so called deposition velocity (CHAMBERLAIN [3]). The physical basis of (8) is that the downward flux of a certain pollutant from a level at some height above the ground equals the product of a deposition velocity, which is mainly a function of the type of surface, and the concentration at the level. A value of 0.01 m/s was used for ν_d . This value can be regarded as an average of SO₂ over land (GARLAND *et al.* [7], OECD [12]).

At the upper boundary of the mixing layer (determined as described under point 3.2.1) the $K(z)$ value was set at zero. This corresponds to an assumption of zero flux of pollutant through this upper boundary.

For the determination of the vertical concentration profile, methods roughly similar to the one applied here have also been used earlier in investigations on the long range transport of air pollutants (BOLIN *et al.* [1], DRAXLER *et al.* [4]).

Figure 3 shows the results of two profile calculations, on the one hand using the diffusion equation and on the other a common source depletion method (VAN D. HOVEN [21]). The main difference between the results of the two

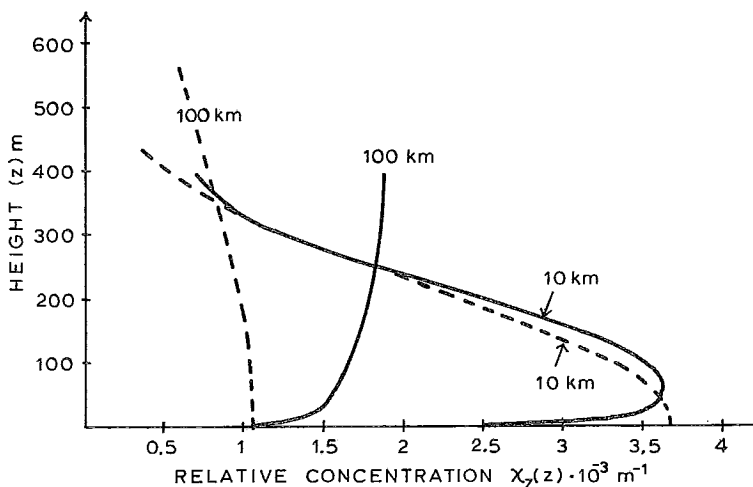


Figure 3. Vertical relative concentration profiles $\chi_z(z)$ for distances of 10 and 100 km from a release at 100 m height calculated with the gradient-transfer method (solid lines) and the Gaussian model (dashed lines). The profiles are for neutral stratification, wind speed 5 m/s and a deposition velocity of 0.01 m/s.

methods is in the concentration distribution close to the ground. The Gaussian method gives slightly increasing concentration values towards the ground, while according to the diffusion model the concentration is considerably lower near the ground than higher up. At least purely qualitatively the results of the diffusion model agree much better with observations (e.g. PASQUILL [13], page 230; OECD [12]).

It is also interesting to study the ground level values calculated for different release heights as a function of time (Figure 4). It may be observed that after long transport times the ground level concentration depends relatively little on the release height. However, the results in Figure 4 apply to neutral stratification; in a stable situation, the influence of the release height will be longer lasting.

In this investigation only the neutral $K(z)$ values have been applied in determining $\chi_z(z)$ for distant sources, since had the $K(z)$ values for stable stratification been used, there would have been an extremely sharp decrease in concentration towards the ground, and this was seen as unrealistic due to the rough terrain and the relatively strong wind even during the night.

The $\chi_z(z)$ value at the height of the sampler was used in the concentration calculations.

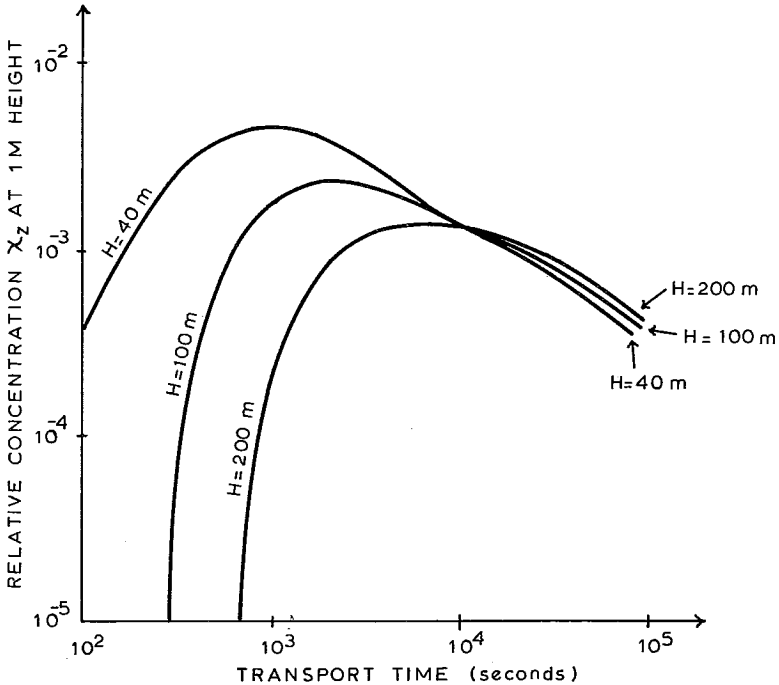


Figure 4. Calculated χ_z values for the height of 1 m above the ground as a function of transport time for release heights of 40, 100 and 200 m; stratification neutral and deposition velocity 0.01 m/s.

3.3 Calculation of maximum domestic contributions

An estimate was made of the biggest possible contributions from domestic sources to the sulfur concentrations measured. Values deliberately underestimating the diffusion were used for these estimations, and accordingly, the following modifications were made to the calculations:

- for low sources (effective chimney height < 100 m) the mean wind velocity \bar{u} in formula (2) was estimated for a level two meter above the ground, instead of for the emission height as in the main calculation
- for sources with an effective chimney height of 100 m or more, the wind velocity estimated for a height of 20 m was used for \bar{u}
- the $\sigma_y(x)$ values in (2) were taken directly from the »Turner Workbook» curves, without corrections for sampling time and wind shear
- for sources nearer than 10 km from the measurement point, the greatest value of the vertical distribution $\chi_z(z)$ was used, corresponding to the value at the plume axis

- for transport distances greater than 10 km the SO₂ released was assumed to be homogeneously distributed in the vertical within the mixing layer, estimated from the Jokioinen and Luonetjärvi soundings as described under point 3.2.1
- the effect of the deposition was neglected

The first and the last of the above modifications brought about the greatest increase in the contributions. For sources located more than 200 km from the measurement point, the last two assumptions alone gave up to four time increased concentrations.

4. Emission values and release heights

The domestic sulfur emissions were estimated separately for heating, industry and pulp mills. Sulfur emissions due to traffic and from natural sources were not taken into account, as they were considered to be of minor significance. The emissions due to heating were calculated based on statistics on building volumes in different rural districts and in towns. Using these volume values, the annual consumption of oil was estimated according to the rule that heating 1 m³ requires 8.2 l oil. (This value was received from the Finnish Property Owner's Association.) Heavy oil, which has a greater sulfur content than the light oil, is used to some extent in apartment blocks. However this is compensated by heating using electricity and heating with firewood, which has a very low sulfur content.

The emissions from industry and power plants were estimated according to statistics on consumption of fossil fuels. For the different types of fuels the following sulfur contents were used:

anthracite	0.018 kg S/kg fuel
duff	0.0079 »
»grate« coal	0.0068 »
coke	0.0035 »
light oil	0.0042 »
heavy oil	0.0255 »

These average values were obtained from the main importers.

The process emissions from sulfate and sulfite pulp mills were estimated by using statistics on the quantity of pulp produced and from factors giving the corresponding amount of sulfur released into the air. These factors for the ratio sulfur emission/pulp produced were from an industrial research company (EKONO).

The process emission values for oil refineries have been received from Neste Oy, the state oil company.

Since March 25, 1973 was a Sunday, the industrial emissions on that day were

smaller than on working days. According to an estimate from the Confederation of Finnish Industries, 2/5 of the industrial concerns were working on this day. The emission values for industry, excluding power plants, pulp mills and oil refineries, were reduced correspondingly.

For emissions greater than about 100 g SO₂/s an effort was made to use the exact locality of the emission. Smaller emissions were added and treated as one emission located at the center of the district in question. However, sources nearer than 5 km were treated individually, and in particular emissions closer than 500 m from the measurement points were investigated carefully in order to get their exact emission value.

Estimated average values, according to the scheme below, were used for the effective release height, *i.e.* the estimated sum of the physical stack height and the plume rise.

Type of source:	Release height used:
small houses and apartment blocks houses in rural districts	20 m
apartment blocks in towns small factories	40 m
ordinary factories	100 m
big factories, pulp mills and power plants	200 m

5. Results

Table 2 shows the most probable domestic contributions to the sulfur concentrations at the different measurement stations. Table 3 presents the corresponding results for the maximum contributions. Finally, in Table 4 the calculated daily mean values are compared with the measured values. The daily mean values are calculated as an arithmetic mean from the trajectory values at 08, 11, 14, 17, 20, 23, 02, 05 and 08 LCT, each representing a 3-hour period with the exception of the 08 trajectories, which represented 1½ hours each.

The individual trajectories for the different measurement points are shown in Figures 5–7. It may be noted that the trajectories are relatively close together and obviously there would have been no need for a shorter time interval than that used, *i.e.* 3 hours between the trajectory calculations. A rough estimate of the influence of possible errors in the direction of the trajectories is given in the next section.

The results (Table 4) show, that domestic contributions are small compared with the measured values and the main part of the sulfur must have been of foreign

Table 2. The calculated SO₂-concentrations (µg/m³) due to domestic sources for the trajectories arriving at the different stations at 08, 11, 14, 17, 20, 23, 02, 05 and 08 LCT, March 25–26, 1973. The last column gives the mean for the 24-hour period 08–08 LCT.

Station	Time of arrival for the trajectories									calc. 24-hour mean
	March 25						March 26			
	08	11	14	17	20	23	02	05	08	
Jokioinen	2.8	2.3	1.8	1.8	2.3	1.9	2.2	0.6	0.8	1.8
Puumala	2.5	10.6	6.3	4.3	1.4	6.2	2.5	6.2	17.1	5.9
Ähtäri	5.1	6.5	1.3	3.3	7.0	9.2	7.3	10.3	8.4	6.5

Table 3. Estimated maximum SO₂-concentrations (µg/m³) due to domestic sources for March 25–26, 1973.

Station	Time of arrival for the trajectories									calc. 24-hour mean
	March 25						March 26			
	08	11	14	17	20	23	02	05	08	
Jokioinen	12.7	4.8	7.7	6.6	9.5	7.6	11.2	2.9	4.3	7.4
Puumala	23.8	72.3	12.8	14.4	1.8	8.8	8.4	33.0	27.5	22.1
Ähtäri	30.4	22.7	5.8	9.3	27.1	58.9	48.0	87.7	87.6	39.8

Table 4. The calculated most probable and maximum domestic SO₂-concentration contributions for the different stations for March 25–26, 1973, compared with the measured values. The last column gives the measured SO₄-values for the same time period.

Station	Daily mean (08–08) LCT:		Measured	Difference:		Measured
	most probable domestic SO ₂ -cont: (C ₁)	estimated maximum SO ₂ -cont: (C ₂)	daily mean SO ₂ (C _M)	C _M -C ₁	C _M -C ₂	SO ₄ -value
Jokioinen	2 µg/m ³	7 µg/m ³	14 µg/m ³	12 µg/m ³	7 µg/m ³	7 µg/m ³
Puumala	6 »	22 »	30 »	24 »	8 »	10 »
Ähtäri	7 »	40 »	49 »	42 »	9 »	27 »

origin. Not even the estimated maximum possible contribution could explain the values measured. It is interesting to note that, according to the calculations, the most probable foreign contribution is only 12 µg/m³ at Jokioinen but 42 µg/m³ at Ähtäri. Since the main airstream was from the south-west it is a little surprising to find higher foreign contributions at Ähtäri, since it is located north of Jokioinen.

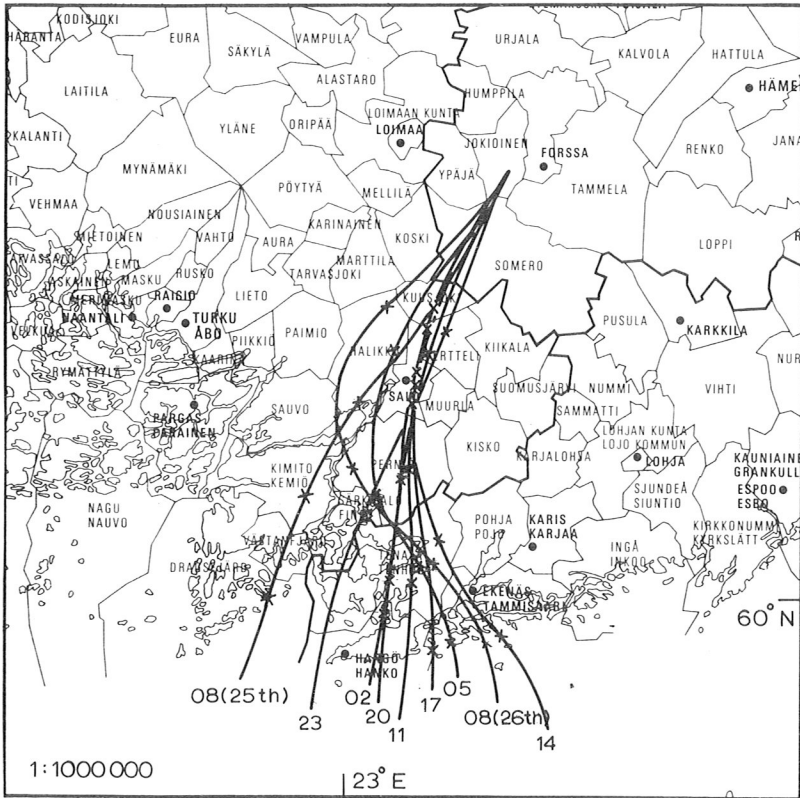


Figure 5. Calculated backwards trajectories from Jokioinen for March 25–26, 1973 08–08 LCT. The stars plotted on the trajectories indicate 2h transport times. The numbers at the end of the trajectories show the time at which the trajectory started (backwards in time) from the measurement point.

If there were no major errors in the measured values^{*}, and the estimated domestic contributions are correctly calculated, there are only two possible explanations: either the vertical stratification of the sulfur was different at two stations, or the foreign sulfur was, in spite of the long transport times, unevenly distributed horizontally. It is difficult to determine which is, in fact, the case. The terrain at Ähtäri is rougher than at Jokioinen and this could have caused a more effective mixing in the vertical, bringing the sulfur down from higher levels; aircraft meas-

^{*} Unfortunately, there is no way of checking measured values afterwards, but the persons responsible for the measurements are of the opinion that the measurement reliability is relatively good for concentrations greater than about $10 \mu\text{g}/\text{m}^3$.

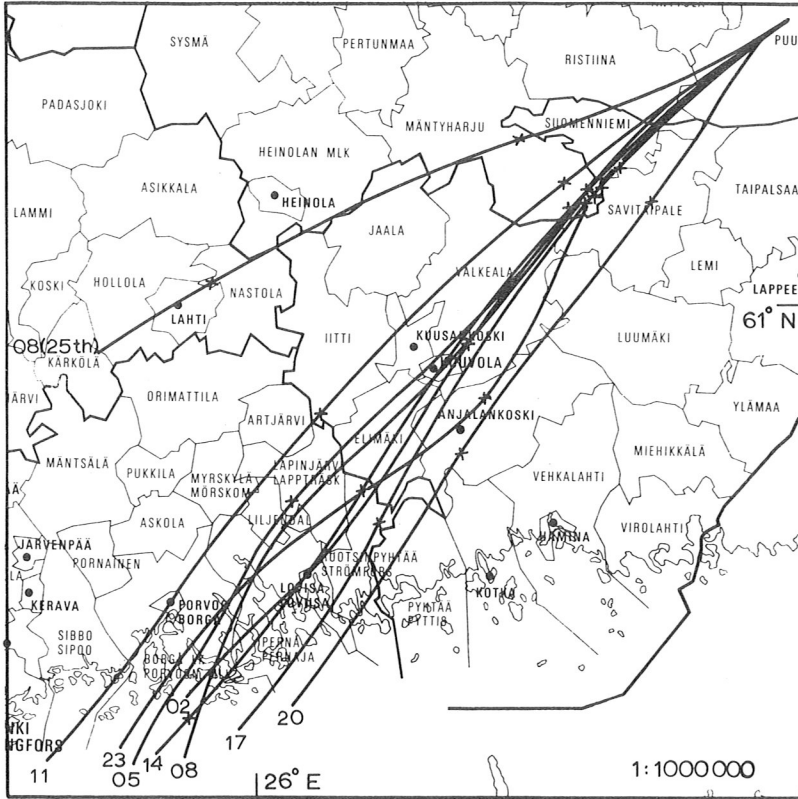


Figure 6. As in Figure, but for Puumala.

Measurements have shown that far from the source the pollutant is mainly transported at a level of several hundred meters above the ground (e.g. PASQUILL [13] page 230, SMITH *et al.* [19]). On the other hand some papers (WENDELL *et al.* [22]; GILLANI *et al.* [8]) have presented results which indicate that air pollutants may be plume-distributed even very long distances from the sources.

The last column of Table 4 gives the measured SO_4 values. Since the transformation rate from SO_2 to SO_4 is relatively small, of the order of $3.5 \cdot 10^{-6} \text{ s}^{-1}$ (OECD [12]), most of the sulfate must have been of foreign origin. Only for Ähtäri were transport times from some sources long enough for a more substantial oxidation. Using the above transformation rate, the domestic contribution of SO_4 was calculated to be about $1.5 \mu\text{g}/\text{m}^3$.

For comparison it is interesting to note that the measured values at Jomala in the Åland Islands were $27 \mu\text{g}/\text{m}^3$ for SO_2 and $21 \mu\text{g}/\text{m}^3$ for SO_4 .

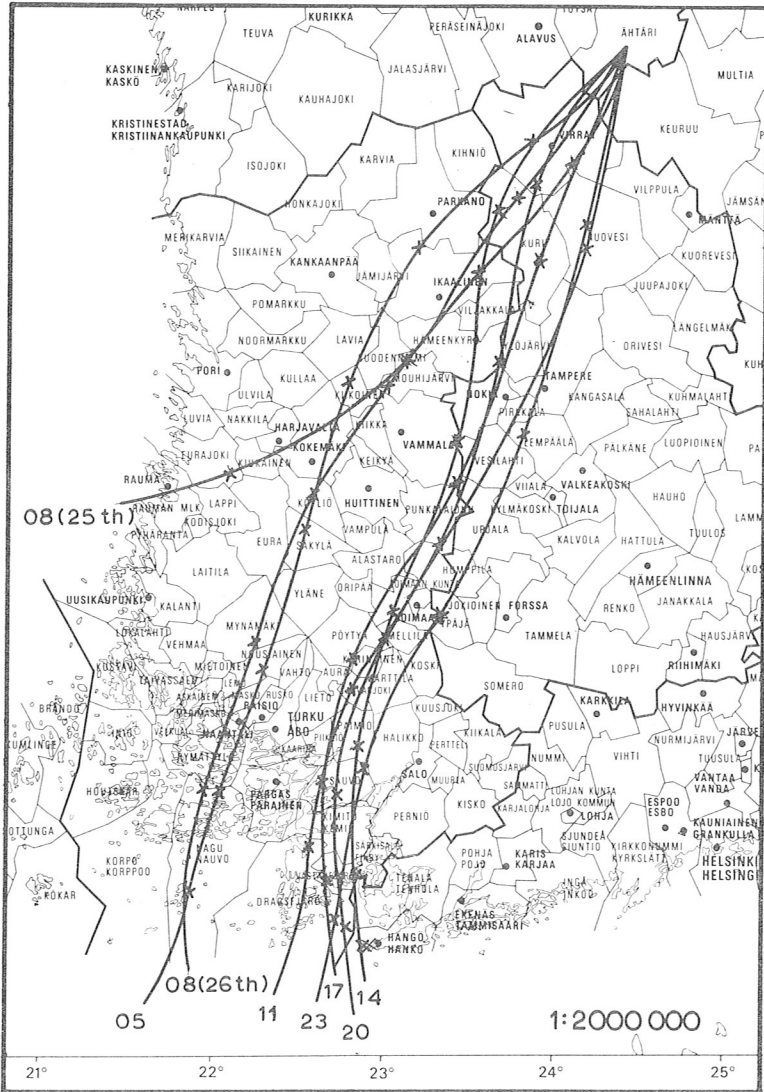


Figure 7. As in Figure 5, but for Åhtäri.

6. Uncertainty analysis

Possible sources of error in the calculated domestic contributions were:

- errors in the emission values

- wrong trajectories
- incorrect diffusion estimates

The emission values can be considered relatively reliable. Possibly the emissions were overestimated in some cases, especially for individual industries. Throughout underestimated values have hardly been used and the calculated domestic contributions are, due to errors in the emissions, often a little overestimated.

The errors in the calculated trajectories are difficult to estimate. However, some upper limits can be given. During the 24-hour period studied the windfield over Finland was relatively homogeneous. Additionally the influence of possible errors in the individual wind observations was reduced by using three observations for the horizontal wind interpolation. One quite subjective estimate is that, in general, the errors in wind direction analysed were less than 10 degrees.

In calculating the trajectories it was rather difficult to decide to what extent the wind shear should be taken into account, i.e. what was the most representative level for the plume trajectories. However, errors in the trajectories due to the use of an incorrect transport level in the calculations occurred almost only during night, when the wind shear was more significant. A reasonable estimate is that the real trajectory lay somewhere between the trajectory formed by the surface winds in which the shear is not taken into account and the trajectory for which the shear up to the top of the mixing layer was fully considered. According to this the errors in the direction of trajectories could have been maximally about 15 degrees. (The maximum shear in direction was about 30 degrees and about half this value was used in calculating the trajectories.)

The influence of possible errors in the trajectories is entirely dependent on the distribution of emissions. A deviation of up to 30 degrees in the direction of the calculated trajectories did not give significantly greater domestic contributions at Jokioinen. For Puumala, a displacement throughout of no more than 5 degrees to the west of the trajectories would have increased the domestic contribution of SO_2 by about $12 \mu\text{g}/\text{m}^3$ over the 24-hour period, since the influence of the big Kuusankoski emissions and the emissions from Sköldvik oil refinery would in that case have been much stronger. In the results for Ähtäri a displacement of about 5 degrees to the east would have given somewhat greater domestic values due to the Tampere area emissions. In stable stratification and with a surface wind velocity of 3 m/s the influence of the Naantali sources at Ähtäri is about $20 \mu\text{g}/\text{m}^3$ for a sampling time of 10 minutes and if the plume is going straight over. The corresponding average influence for a sampling time of 3 hours is only about half as much, due to the effect of meandering. If the daytime trajectories from Ähtäri had gone nearer Naantali, this would in every case have given a significant increase in the domestic contributions.

Since the wind velocity increased rapidly with height, the height at which \bar{u} in (2) was taken was significant. The height of the release can be considered as a natural choice. However, in studying ground level concentrations it would also be justifiable to use a lower level than the release height for determining \bar{u} . This would have given somewhat decreased dilution in the mean wind direction and correspondingly increased calculated domestic concentration contributions.

It is interesting to note that the influence of variations in the $\sigma_y(x)$'s of the horizontal dispersion is highly dependent on the y value in (2), *i.e.* on how far the source is located from the trajectory. From the Gaussian distribution it can be obtained that the greatest sensitivity to errors in σ_y occurs when $y = \sigma_y(x)$. According to this relation and to the distribution of sources, it was found that errors in $\sigma_y(x)$ would have had relatively little influence on the concentration contributions from the single trajectories.

The method of estimating the vertical diffusion value $\chi_z(z)$ at ground level obviously gave somewhat overestimated contributions under stable conditions. In some cases the ground level $\chi_z(z)$ values may even be greatly overestimated, but it is hardly likely that they were significantly too small. In a relatively well-mixed boundary layer, limited by an upper inversion, there is obviously no reason for the ground level concentration to be higher than the mean over the mixing layer, at least not when the source is elevated. In a stable stratification the plume from an elevated source is mainly transported at the release level, and a situation in which the concentration is at its greatest near the ground is most unlikely.

7. A separate calculation

It may be worth mentioning, quite briefly, another calculation in which the same method was used for calculating domestic contributions to Jokioinen over a period of 24 hours (March 23–24, 1973). On this occasion the measured value was $15 \mu\text{g}/\text{m}^3$, but according to model calculations (NORDLUND [11]) no significant long-range transport should have occurred.

The trajectories for this day are shown in Figure 8, and the corresponding estimated domestic concentration contributions are given in Table 5.

It is interesting to note the wide spread of the trajectories, which among other things indicates that for this day only one or two trajectories would not have been enough to determine the sources of the concentrations observed.

The high values for the times 08 and 14 (23rd) and 08 (24th) are, according to the calculations, based mainly on local sources at Jokioinen. The small difference of $2.5 \mu\text{g}/\text{m}^3$ SO_2 between the calculated domestic contribution and the measured

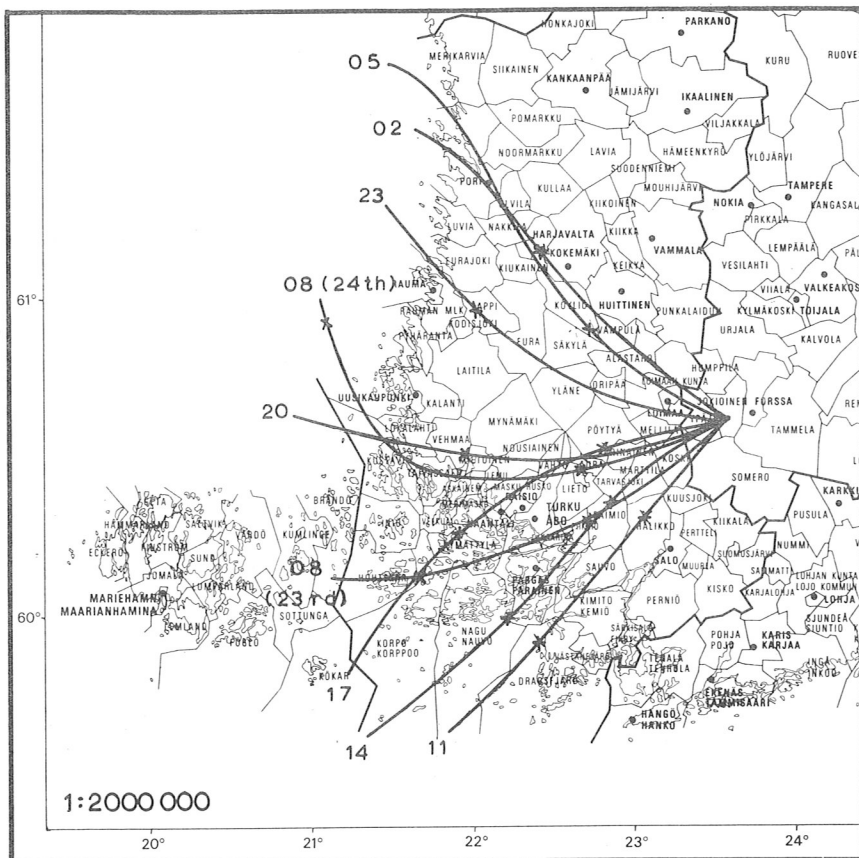


Figure 8. As in Figure 5, but for the period March 23–24, 1973, 08–08 LCT.

Table 5. The calculated domestic SO₂ concentration contribution ($\mu\text{g}/\text{m}^3$) at Jokioinen for March 23–24, 1973 compared with the measured value. The calculated contributions are given separately for the different trajectories and as a mean for the whole 24-hour period.

Station	Arriving time for the trajectory									calc. 24-hour mean	measured 24-hour mean
	March 23			March 24			02	05	08		
Jokioinen	08	11	14	17	20	23	6.7	6.8	36.5	12.5	15

value is within the limits of calculation errors. Contributions from Sweden and the global background level of SO_2 , which is of the order of some $\mu\text{g}/\text{m}^3$ (ERIKSSON [5]), could also have caused the difference.

8. Conclusions

According to the results the high sulfur concentration values measured at Jokioinen, Puumala and Ähtäri on March 25–26, 1973 cannot be attributed to domestic sources. A considerable part must have been of foreign origin.

The most probable values for the foreign contribution to the ground level sulfur dioxide concentrations were $12 \mu\text{g}/\text{m}^3$ at Jokioinen, $24 \mu\text{g}/\text{m}^3$ at Puumala and $42 \mu\text{g}/\text{m}^3$ at Ähtäri. For sulfate, almost the entire measured concentrations of $7 \mu\text{g}/\text{m}^3$ at Jokioinen, $10 \mu\text{g}/\text{m}^3$ at Puumala and $27 \mu\text{g}/\text{m}^3$ at Ähtäri were estimated to be of foreign origin. Only for Ähtäri the domestic influence was estimated to be greater than $1 \mu\text{g}/\text{m}^3$, and even there only about $1.5 \mu\text{g}/\text{m}^3$, the foreign contribution, at least for Ähtäri and Puumala, must be considered very high compared with long-term means and with the global background sulfur concentrations of »clean air».

A weakness in the present investigation is, of course, that it covers only a 24-hour period. However, the principles of the method presented for calculating pollutant concentration contributions from local and more distant sources will be used in future investigations, too.

On the basis of the present investigation it can, once again, be pointed out that under certain weather conditions considerable long distance transport of sulfur, and probably also of other airborne pollutants, occurs.

Acknowledgements: For helpful discussions and comments the author wishes to thank Mrs. Pia Bremer of the Finnish Meteorological Institute. Many thanks are also due to the FINNCELL company for giving help in evaluating the emissions from pulp mills. Finally the author wishes to thank Mr. M. Mannermaa and Mrs. E.-L. Honkanen of the Finnish Meteorological Institute for excellent technical assistance.

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APPENDIX 1 Emission values used SO₂ on working days 1973) for major domestic sources influencing the concentrations at the measurement points on March 25–26, 1973.

Measurement point:

Jokioinen

Source	approx. distance	emission value SO ₂
Agricultural research centre*	0.3 km	0.027 g/s
Jokioinen syrup-factory*	0.6 »	2.15 »
»Jokioisten Kartanot»-estate	1.1 »	0.054 »
Jokioinen centre	1.2 »	0.084 »
Fiskars-factory	1.2 »	0.15 »
Salo town	50 »	26.6 »
Kemiö district	75 »	4.2 »

Puumala

Kuusankoski pulp mills	110 »	562 »
Kuusankoski other industries	110 »	285 »
Heinola town	115 »	89 »
Myllykoski industries	120 »	163 »
Anjalankoski industries	120 »	25 »
Lahti town and industries	150 »	231 »
Kotka-Karhula-Kymi district	150 »	630 »
Sköldvik oil refinery	200 »	1470 »
Tolkis pulp mill and Porvoo district industries	200 »	238 »

Ähtäri

»Salonranta» house	0.15 »	0.0011»
»Inha» factory	4.0 »	0.022 »
Virrat disctrict	40 »	4.3 »
Lielähti pulp mill	120 »	287 »
Tampere town and industries	120 »	470 »
Nokia district, industries	125 »	367 »
Harjavalta industries	200 »	168 »
Rauma town and industries	220 »	311 »
Turku town and industries	260 »	198 »
Raisio district and industries	260 »	33 »
Naantali, town and industries	265 »	442 »
Naantali oil refinery	265 »	672 »
Pargas, district and industries	265 »	126 »

*Influencing only the calculation for March 23–24.