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ESTIMATING CONTRIBUTIONS OF SOURCE AREAS TO THE MEASURED YEARLY AVERAGE SO_2-CONCENTRATION FIELD IN THE NETHERLANDS BY DISPERSION-MODEL PARAMETER-OPTIMIZATION

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Abstract – The contributions of major source areas inside and outside the Netherlands to the SO_2-concentrations throughout the country (200 × 300 km), are estimated by optimizing the parameters of a dispersion model with respect to the measurement results of a SO_2-monitoring network of 99 stations. The estimations are made both for hourly average and for yearly average fields. The optimized model, which accounts for deposition losses during the mesoscale transport, describes 85% of the spatial variance of the yearly average concentration field. Only 30% of the overall yearly average SO_2-concentration in the Netherlands could be ascribed to inland sources.

INTRODUCTION

The analysis of data, obtained from air pollution monitoring networks generally is directed to either a pure statistical description of the space–time behaviour of the pollutant under study or the validation of diffusion models. Especially with respect to the testing of public health criteria, preferably statistical methods are used, involving no assumptions about the dispersion process and only a few about the statistical properties of the concentration field. In order to enable a physical interpretation of such descriptive results, a minimum spatial sampling resolution is required, so that a (statistical) extrapolation beyond the spatial arguments of the monitoring sites can be made, followed by the reconstruction of the underlying continuous concentration field.

In previous work (van Egmond *et al.*, 1978) the mesoscale transport of sulphurdioxide in the Netherlands was studied on the basis of such reconstructed concentration fields and additionally obtained spatial profiles of vertically integrated concentration (gas-burden). The concentration field was derived from the measurement results of a nationwide (200 × 300 km) monitoring network with interstation distances of about 20 km. For a number of case studies, substantial transports of SO_2 from the major source areas inside (Rijnmond) and outside the country (e.g. Ruhr–Germany, Antwerpen–Belgium) were found, indicating that throughout the Netherlands the current maximum concentrations, as given by the 98-percentile levels, are affected strongly by these transports. Due to the use of no-sulphur containing natural gas for domestic heating, the spatial SO_2-concentration gradients are small and the resolution of the network is sufficient.

As the natural gas resources will be exhausted within a few decades the use of gas by large consumers such as power plants, will be restricted on a short term and sulphur containing fuels like oil and coal will be reintroduced, giving rise to important SO_2 emissions. For arriving at a rational fuel allocation and emission control-strategy, diffusion models will be used to estimate the long term average concentrations of existing and future sources. Within this framework the contributions of the mesoscale transports to the current average SO_2-levels have to be determined as much as possible based on measurement results and as little as possible based on hardly justified assumptions about the complex physical transport process. However, in order to decompose the concentration field of yearly average concentrations into separate contributions of the individual (industrial) source areas, some minimum assumptions about the physical transport process have to be made. For this reliable information about the source-strengths of these areas and detailed meteorological information is required, but not available. Moreover, the previous case-studies indicated that a substantial part of the transport takes place above the ground based mixing-layer, not affecting the ground concentration over large distances from the source areas.

To overcome these complications an approach was chosen in which the parameters of a simple dispersion model are optimized with respect to the actual measurement results of the SO_2-network. As the influence of wind-direction fluctuations does not seem to be large, a Gaussian type model was used, which accounts for the deposition of SO_2 over mesoscale transport distances (30–300 km).

The idea of fitting analytical dispersion models to sparse data on air quality was exploited by Heimbach

and Sasaki (1975) for the computation of concentration fields in cases where the network resolution is not sufficient for the direct application of statistical interpolation techniques. Gustavson (1975) gives a numerical experiment for an urban area, arriving at directly applicable interpolation formulas. In the present study the technique will be directed to the estimation of unknown effective source strengths and the computation of individual contributions to the measured mesoscale concentration fields. The method forms a link between aforementioned statistical data-analysis techniques and model validation procedures by relating the model description as directly as possible to the measured concentrations.

DATA

The computations are based on the hourly average values of SO_2-concentrations obtained from 99 baseline-grid stations of the Dutch nationwide automated monitoring system, covering the total area of the Netherlands (200 × 300 km). The interstation distance is about 20 km. The SO_2-concentrations are measured by means of Philips PW 9755 monitors, which are calibrated every 12 h and corrected for zero drift every 3 h, assuring good interstation comparability. In general, the baseline-grid stations are situated outside the direct influence of local SO_2-sources.

DISPERSION MODEL

The data-analysis method will be applied both to hourly concentration fields and to fields of average concentrations per windspeed–wind direction category. The only function of the dispersion model is to describe the spatial interrelationships in the concentration field, due the transport from assumed source areas. Consequently, the model can be rather simple, but nevertheless should account for deposition and decay during transport.

As the concentrations are to be calculated in the first place for mesoscale distances (30–300 km) from the source areas, vertical dispersion is not taken into account and in the direct vicinity of these areas a correction factor will be introduced. As a mixing height climatology is not available at the present state, it has to be assumed that the effects of deposition at various heights add up to the effect of a single effective, but unknown mixing height. Formally, the weighted sum of the negative exponential functions, by which the concentration change by deposition in the transport direction is described, does not result in another negative exponential function. However, from several numerical experiments, in which the profiles for mixing heights in the range from 300–1000 m were averaged, it was concluded that no substantial errors were made in approaching the resulting profile by a simple negative exponential function and consequently by a single "effective" mixing height.

The "slender plume" approach will not be followed as the computations are performed for individual concentration fields and discontinuities in the modelled fields are to be avoided.

These considerations lead to the following simplified (Gaussian) model:

$$C(x,y) = \frac{Q}{\sqrt{2\pi}\,uL} \frac{\exp\left[-\frac{1}{2}\left(\frac{y}{\sigma_y}\right)^2\right]}{\sigma_y} \exp\left[\frac{-v_g x}{uL}\right], \tag{1}$$

where;

L = the mixing height, considered to be an effective value such that the third factor approaches the average of a large number of individual negative exponential functions,

Q = the effective source strength, which is the emission fraction transported in the ground based mixing layer with height L, depending on the complex emission height and plume-rise conditions within the source area,

u = the average windspeed in the mixing layer,

v_g = the deposition velocity, accounting for all loss processes which are proportional to the SO_2-concentration,

x and y = positional coordinates, relative to the source,

σ_y = the horizontal dispersion parameter; by Pasquill (1974) σ_y is given by $\sigma_y = px^{0.89}$, where p depends on the atmospheric stability,

$C(x,y)$ = the concentration at position x, y.

The area sources are treated as virtual point-sources according to the method proposed by Martin (1971). The virtual distance x_0 is chosen such that the source area diameter D equals the plume width, i.e.

$$4\sigma_y = D.$$

Averaging over a wind-direction sector of 30° is achieved by averaging over the plume cross-section. For this purpose the value of $\exp[-\frac{1}{2}(y/\sigma_y)^2]$ at crosswind-position y is replaced by the average value over the interval $y \pm \frac{1}{2}(x - x_0)\frac{1}{6}\pi$, which can be obtained directly from the tabulated values of the error function, thus evading an increase in computation time. In this two-dimensional model, unrealistic overestimations occur in the direct vicinity of the source areas, where the ground concentrations primarily are determined by the dispersion conditions in the third dimension, i.e. the effective plume height and the vertical dispersion σ_z. To account for this nearby situations in which the elevated plume only partly affects the ground concentrations, the simple correction factor $\sigma_y(x)/\sigma_y(15)$ is introduced, in which $\sigma_y(15)$ is the dispersion coefficient at 15 km from the virtual point source. The distance of 15 km is chosen as

the distance at which $\sigma_z/L = 0.9$ at stability class C according to Singer and Smith (1966). Equation 1 may be written as:

$$C(x, y) = Qa(x, y), \tag{2}$$

in which $a(x, y)$ comprises the non-linear factors of the model and can be considered as the transfer-coefficient between emission (Q) and emission (C). Now $a(x, y)$ is a function of only two parameters:

p = the coefficient in the function $\sigma_y = px^{0.89}$ and

uL = the product of windspeed and mixing height. As the deposition velocity v_g is assumed to be constant over space and time and as such set to 0.01 m s^{-1}, the only variable parameter in the exponent v_g/uL is uL. The quantity $\sqrt{2\pi}\,uL$ is part of the linear factor in the model and only affects the absolute value of Q.

OPTIMIZATION OF PARAMETERS

The concentration C_i at monitoring station i is considered to be sum of n individual contributions C_{ij} of sources j, with effective strength Q_j, plus a residual concentration E_i, containing both the measurement and sampling error, and the effect of unknown (local) sources:

$$C_i = \sum_{j=1}^{n} C_{ij} + E_i = \sum_{j=1}^{n} Q_j a_{ij} + E_i.$$

In matrix notation:

$$C = AQ + E$$

where:

C = the vector of measurement concentrations at m stations,

A = the $m \times n$ matrix of transfer coefficients from n sources to m monitoring stations,

Q = the vector of n effective source strengths,

E = the vector of m residual concentrations.

In matrix A a unit column vector accounts for a constant (space-independent) background level. This level will be treated as a normal source.

Now the unknown quantities are estimated by means of the ordinary least squares method:

$$0 = \frac{\delta(C - AQ)'(C - AQ)}{\delta Q} = \frac{\delta E'E}{\delta Q} = -2A'C + 2A'AQ,$$

from which

$$\hat{Q} = (A'A)^{-1}A'C$$

The effectiveness with which the model describes the original (measured) concentration field is evaluated by the parameters:

s = the standard error of estimate, computed as $s = \sqrt{E'E/(m - n - 1)}$,

R^2 = the fraction of the spatial variance (with respect to the spatial mean) which is explained by the model.

Under the assumption of normally distributed residuals, confidence intervals for the estimates $\hat{Q}$ are given by the variances of $\hat{Q}$, obtained as the diagonal elements of the matrix $(A'A)^{-1}\sigma^2$, in which σ^2 is estimated by s^2.

The least squares procedure optimizes the model for given values of the parameters p and uL. A further optimization is now achieved by optimizing the explained variance fraction R^2 as a function of these two parameters. So the least-squares technique is applied for a range of values for p and uL and the field for which R^2 reaches its optimum is decided to be the best description of the measured concentration field in terms of the assumed source areas.

In practice, it has turned out to be impossible to explain the concentration fields from the known source areas only. In the residual concentrations in most cases a large negative gradient i.e. a decrease in concentration in the direction of the wind, was observed over the total measurement area, not having any profile in the crosswind-direction. While the uniform contribution to the background level is ascribed to homogeneously distributed, local SO_2-sources within the Netherlands, the gradient-contribution is expected to originate from remote source areas. To account for this contribution, a line-source at an arbitrary upwind distance was introduced. The existence of significant remote source areas is affirmed by (not-detailed) emission inventarisations given by the OECD (1977) and by the German "Bundesministerium des Innern" (1976).

MODEL RESULTS

Hourly average concentrations

During a major part of the time the wind persists from a given direction and the flow conditions may be considered as stationary. It further seems reasonable to assume that the height of the mixing layer is constant over the entire 200 × 300 km area. Under these assumptions an estimation of the effective source strengths can be made. To each source area, which is expected to affect the measured concentration field, position coordinates and (area-) diameter were assigned. Then the value of R^2 was optimized by varying the parameters p and uL. Sources for which negative or clearly non-significant emissions were estimated, were deleted (in most cases) and the procedure was repeated.

A first example is given for the concentration field of 29 December 1977 – 10.00 h, which is dominated by a transport zone arising from source areas in Belgium and/or France. The concentration field is given in Fig. 1(a), and the corresponding wind-field is presented in Fig. 1(b) in combination with the source locations. At the resulting parameters $u = 7$ m s^{-1}, $L = 200$ m ($uL = 1400$ m^2 s^{-1}) and $\sigma_y = 0.53\ x^{0.89}$, the following

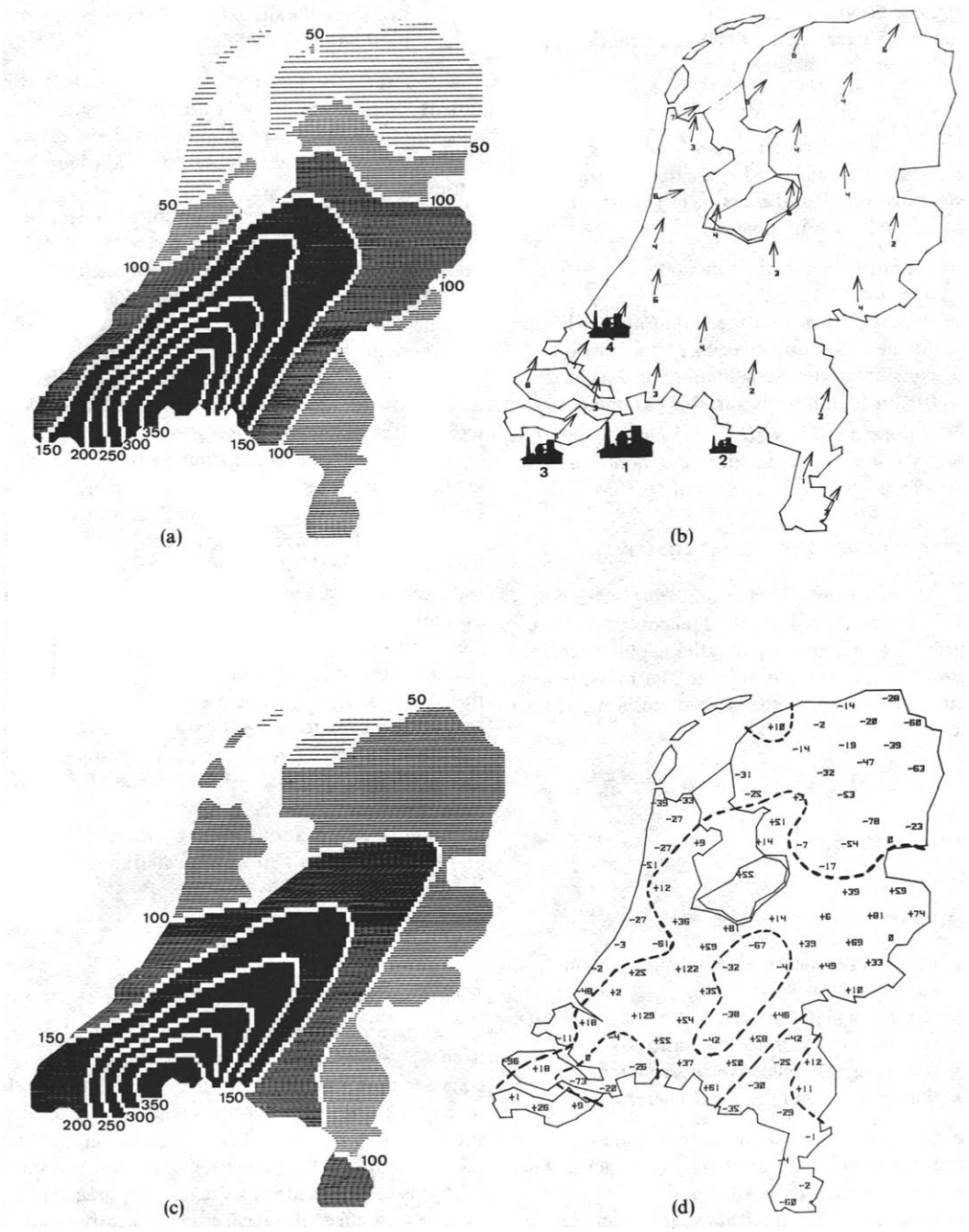

Fig. 1. Measured SO_2-concentration field (a) wind field and source locations (b) modelled concentration field (c) and residual concentrations (d) on 29 December 1977, 10.00 h ($\mu g\ m^{-3}$).

source strengths are estimated from the hourly average concentrations at 99 monitoring stations (standard deviation between brackets):

1. Antwerpen	45 ($\pm$3.6) tons h^{-1} SO_2
2. Balen/Mol	4 ($\pm$3.5)
3. Zelzate/Sas van Gent	42 ($\pm$4.2)
4. Rijnmond	9 ($\pm$3.6).

The contributions from the line source, describing the concentration profile which is due to emissions outside the area covered by the network, i.e. from the Dutch border, range from $\pm 70\ \mu g\ m^{-3}$ in the south west to $\pm 25\ \mu g\ m^{-3}$ in the north-eastern part of the country. In this case this contribution probably includes the emissions near the western part of the French–Belgian border, a few hundreds of kilometers to the south-west. The constant level contribution is estimated as 26 $\mu g\ m^{-3}$. The model explains 81% of the measured spatial variance. The modelled field is given in Fig. 1(c) and the residual concentrations, which are obtained as the difference between measured and modelled concen-

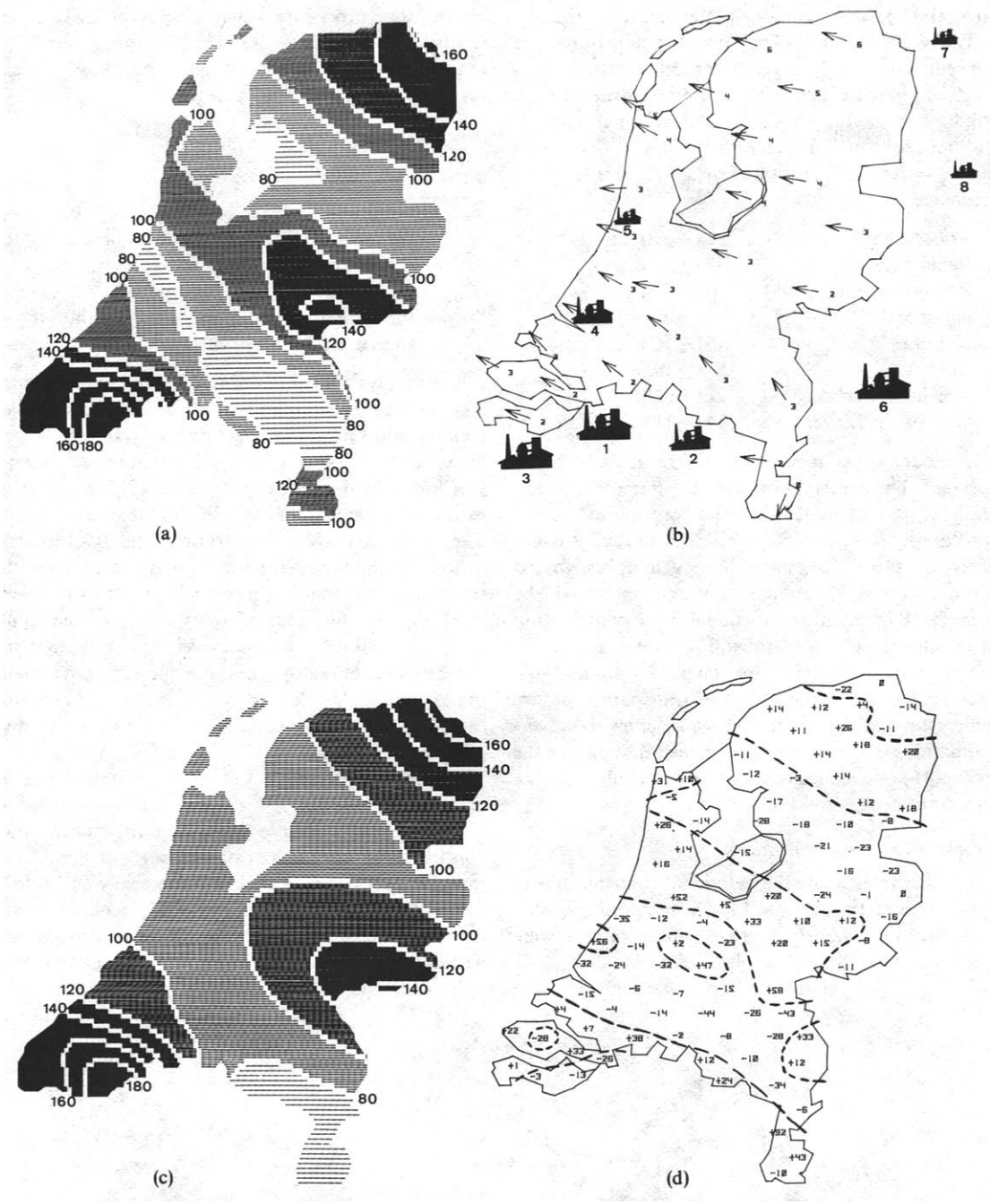

Fig. 2. Measured SO_2-concentration field (a), wind field and source locations (b), modelled concentration field (c) and residual concentrations (d) on 19 January 1978, 13.00 h ($\mu g\ m^{-3}$).

trations, are presented in Fig. 1(d). Areas with positive and negative residuals are indicated by dotted lines, showing that the lack of fit remains due to a systematic discrepancy between measured and modelled results. Apart from small anomalies, the model tends to underestimate over distances up to approx 200 km and to overestimate for larger distances.

The total emission of the Belgian source-areas (91 tons h^{-1}) is in good agreement with the results of mobile transmission measurements where SO_2-transports were found to range from 60 to 90 tons h^{-1} (van Egmond *et al.*, 1978). During 29 December, the mixing-layer height was about 200 m and all SO_2 was transported within this layer, which is considered to be more an exception than a rule. Proof for this limited vertical mixing was obtained from SO_2-measurements in a 200 m meteorological tower in the centre of the country, where high SO_2-levels at 3 and 100 m were recorded from 7.00 to 20.00 h, while at 200 m the concentration level remained below 30 $\mu g\ m^{-3}$, reaching a comparable level of 300 $\mu g\ m^{-3}$ only between 15.00 and 18.00 h, which appeared to be the only time-

interval when the mixing height exceeded 200 m.

The second example of modelling an hourly average concentration field is given for 19 January 1978, 13.00 h; modelled and measured concentration field, wind field, source locations and residual concentrations are given in Fig. 2. At $u = 4\ \mathrm{m\ s^{-1}}$, $L = 500$ m and $\sigma_y = 0.86\ x^{0.89}$, the effective source strengths are estimated as:

1. Antwerpen	22 (±2.8) tons $\mathrm{h^{-1}}$
2. Balen/Mol	35 (±7.4)
3. Zelzate/Sas van Gent	6 (±3.5)
4. Rijnmond	5 (±2.3)
5. IJmond	5 (±5.9)
6. Ruhr	57 (±10.3)
7. Northern Germany	71 (±9.9)
8. Osnabrück/Lingen	34 (±11.6).

The model explains 63% of the measured spatial variance. The contribution from the (remote sources) gradient part is small, while the contribution of the constant level is high (60 $\mu g\ m^{-3}$). Although it is not likely that this is due only to homogenous distributed sources in the Netherlands, this assumption will be made in all cases, and, consequently, the contribution of Dutch sources in general will be overestimated. As can be concluded from the map of residuals, the measured concentrations in the transport zone are underestimated by the model, while elsewhere overestimations occur. This means indeed that part of the transport-zone concentrations are included in the constant background level.

Yearly average concentrations

In order to estimate the individual contributions of major source areas to the 1977 yearly average SO_2-concentrations, hourly average concentrations were classified into windspeed classes of <3, 3–5, 5–8, $>8\ \mathrm{m\ s^{-1}}$ and wind direction classes of 30°. The optimization procedure then was accomplished for the resulting $4 \times 12 = 48$ windcategories and the individual results were summed according to the relative frequencies of the categories:

$$\hat{C}_{ij} = \sum_{k=1}^{48} \hat{C}_{ijk} f_k,$$

where

$\hat{C}_{ijk}$ = the estimated contribution of source area j to monitoring station i in wind-category k,
f_k = the relative frequency in category k,
$\hat{C}_{ij}$ = the estimated yearly average contribution of source area j to station i.

The analysis was performed for 99 monitoring stations and 11 source areas inside and outside the country apart from the larger distance contributions. In most cases only a weak optimum in the R^2-values was found in dependence of p and uL, indicating a restricted sensitivity for the values of these parameters. The yearly average concentration field ($\hat{C}_{ij}$) as described by the model is in very good agreement with the measured concentration field, as may be concluded from Fig. 3. The model describes 85% of the total spatial variability of the measured field and tends to overestimate the concentrations in the northern part of the country slightly. The minor difference in the south-eastern part may be explained by local topography (height-differences).

The contributions of both Dutch and foreign source areas to the 1977 yearly average concentration field is given in Fig. 4. These contributions include the contributions of the constant level part and the (remote sources) gradient-part, respectively. In Table 1 the contributions of the major source categories to the average concentration over the Netherlands is given. The Dutch contribution of 6 $\mu g\ m^{-3}$ amounts to 30% of the overall (space–time) average of 20 $\mu g\ m^{-3}$, while

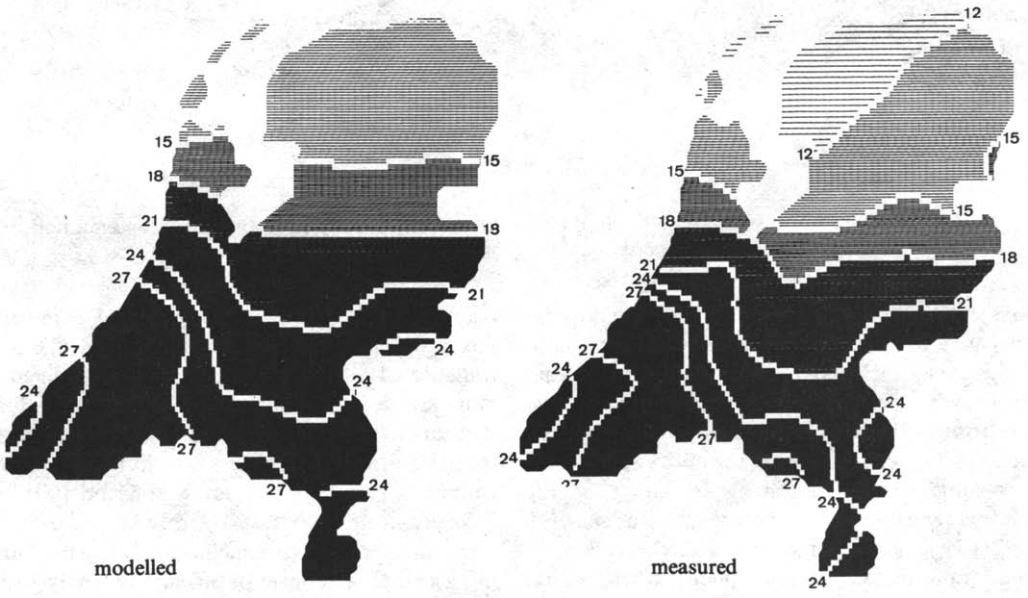

Fig. 3. Measured and modelled yearly average SO_2-concentration field for 1977 in $\mu g\ m^{-3}$.

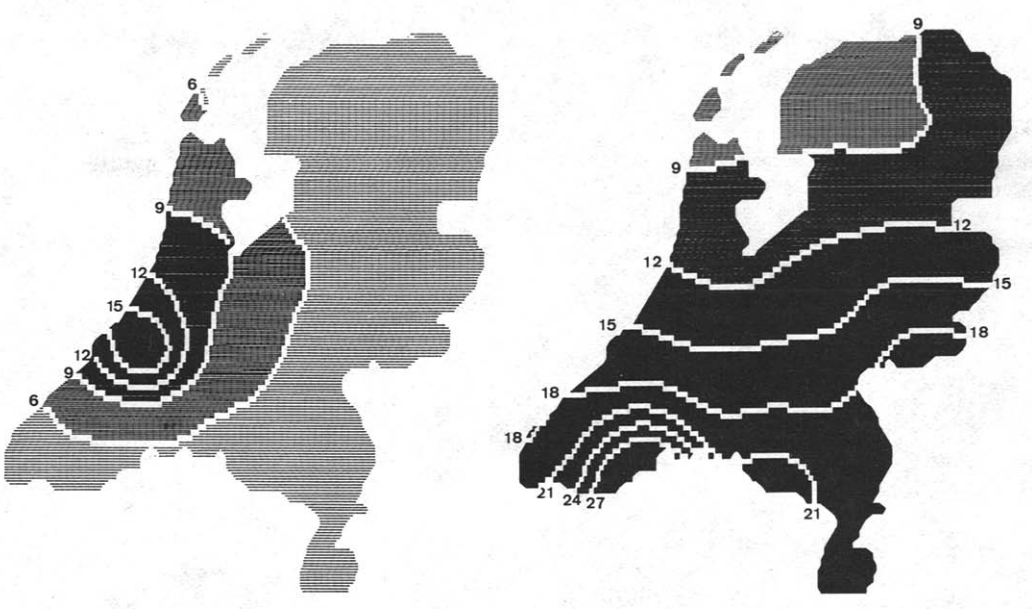

Fig. 4. Total contributions of Dutch and foreign sources to the yearly average SO_2-concentrations over 1977 in $\mu g\ m^{-3}$.

70% should originate from foreign sources. The contributions of remote sources which cannot be allocated spatially, due to the lack of variability in the cross-wind concentration profile, is rather large (7 $\mu g\ m^{-3}$), but it is very unlikely that this contribution should be much smaller. Further, the contribution of small homogeneous distributed Dutch sources is computed as 4 $\mu g\ m^{-3}$, which seems to be a realistic estimate.

Recently, estimations have been made of the contributions of dry depositions of SO_2 from remote source areas to receptor countries. The estimations were made in the LRTAP project (OECD 1977) and by the EURMAP model, developed by Johnson *et al.* (1978) and are based on trajectory methods. The LRTAP model estimates the contributions of Dutch sources to the deposition in the Netherlands over 1974 on 60%, while the EURMAP estimate amounts to 51% for 1973. The statistical method which was presented here, gives an estimate of only 30% and indicates that it is very unlikely that the Dutch contributions should exceed the amount of 50%.

Table 1. Contributions of individual source areas to the overall average SO_2-concentration in the Netherlands over 1977 in $\mu g\ m^{-3}$

Source area	Contribution
Ruhr	3 $\mu g\ m^{-3}$
Osnabrück/Lingen	1
Balen/Mol	1
Antwerpen	2
Other foreign sources	7
Total foreign sources	14 $\mu g\ m^{-3}$
Rijnmond	2
Other Dutch sources	4
Total Dutch sources	6 $\mu g\ m^{-3}$
Overall (space–time) average	20 $\mu g\ m^{-3}$

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