

Long-Range Transport Modeling of Air Pollution Episodes

by Frank A. A. M. de Leeuw* and H. Jetske van Rheineck Leyssius*

The air quality network in the Netherlands is based on a regular grid with an interstation distance of 30 to 40 km. In or close to source areas, a higher station density is realized. Atmospheric transport models form an intrinsic part of the air quality monitoring system. The models are constructed in such a way that only routinely available meteorological input data are needed. The models are applied for interpretation and generalization of the measurements in terms of contributions of source categories. In scenario studies, the potential effect of abatement strategies is explored. The output of the models consists of concentration and deposition fields of NO_x, NO₂, SO₂, sulfate, and nitrate on the scale of the Netherlands or on the scale of northwestern Europe. In the present version the models are not directly suitable to estimate exposures to acidic aerosols; however, in combination with limited aerosol measurements, the model predictions can be used to provide information on the spatial and temporal distribution of acidic aerosols as needed for exposure assessment.

Introduction

In the Netherlands, which are situated downwind of important European source areas, air quality is measured by a nationwide monitoring network covering an area of 200 × 300 km². In order to interpret the observed concentrations, atmospheric transport models, in which the whole chain of emissions, transport, transformation, and removal is described, have been developed. In this paper a short description of the network will be given. The nature of the transport models is discussed. In the discussion, emphasis is on the capability of the models to predict exposures to acidic aerosols.

Monitoring Network

In the Netherlands, the national air quality monitoring network has been operational since 1976. The main objectives of the monitoring program are geographical mapping of pollutant levels for compliance with public health standards, both on the long term and during episodes with high pollution levels; and estimation of source contributions to the observed pollution patterns.

Based on statistical analyses (1) of earlier measure-

ments during the period 1976 to 1985, an optimal configuration of the network was calculated and realized in 1986. The interstation distance in the northern part of the country is 40 km, in the more polluted southern part this is 30 km. In or close to the source areas, a higher density is found (Fig. 1). Additional stations are located in cities on sites that are representative for various traffic situations.

Continuous measurements are performed for the gaseous components sulfur dioxide (SO₂), nitric oxide (NO), nitrogen dioxide (NO₂), ozone (O₃), and carbon monoxide (CO). At a subset of 17 special stations (actually small field laboratories), additional measurements of particulate matter containing heavy metals and organic compounds, rainwater concentrations, and biological effects on indicator plants are performed. To supplement the monitoring network, measurements are made by means of specially equipped vans. These vans are equipped with instruments for concentration measurements and with optical measuring instruments for estimating pollutant fluxes from nearby and remote source areas. Another application is in the study of local pollutant levels near single sources.

In each monitoring station a microcomputer pre-processes the raw measurements. Hourly averaged concentration data are transmitted by dial-up lines to the central acquisition system in Bilthoven. Results of the automatic network are real-time distributed to provincial governments via the PTT Viditel system

*National Institute of Public Health and Environmental Protection, P.O. Box 1, 3720 BA Bilthoven, the Netherlands.

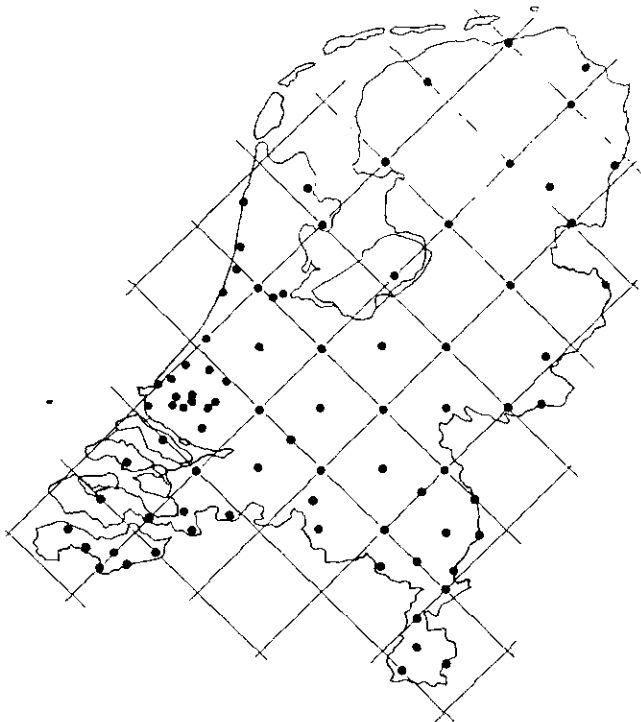


FIGURE 1. Configuration of the air quality monitoring network.

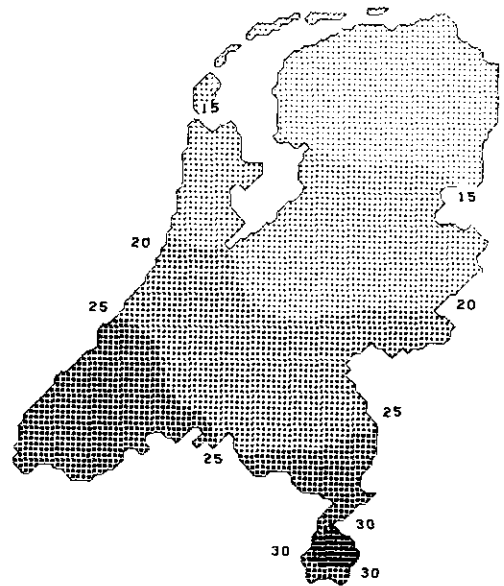


FIGURE 2. Yearly average concentration field of SO_2 ($\mu\text{g}/\text{m}^3$), 1985.

and distributed to a broad public in the form of maps via NOS Teletext.

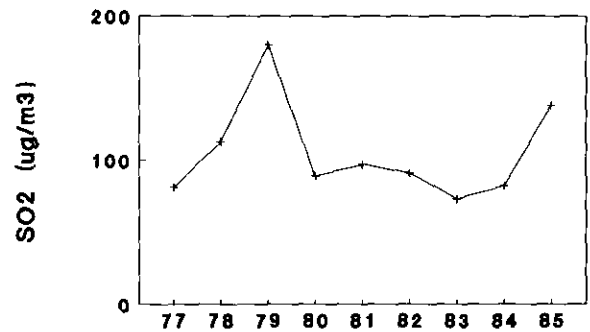
Measurement Results

As an example of the results obtained by the network (2), the spatial distribution of the yearly averaged SO_2 concentrations is given in Figure 2. Figure 3 shows that the 50th percentile values decreased since 1979, reflecting the reduced SO_2 emissions both in the Netherlands and in its direct surroundings. The 98th percentile values show peaks in 1979 and 1985. In both years, episodes with extremely high concentrations occurred during unfavorable meteorological conditions. In Table 1 (2,3) concentrations both on a yearly average base and during episodes are summarized. Note that the concentration of acidic aerosols contributes only a small fraction to the total atmospheric acidity: approximately 30% for yearly averaged concentrations. On a yearly basis, acid sulfates contribute about 20% of total sulfate, during episodes this increases to 30 to 70%.

Transport Models

To explain the observations in the Netherlands in terms of contributions of various source categories and to explore the potential effect of policy scenarios, models have been developed that describe transport, transformation, and deposition. The models are applicable on the scale of the Netherlands and direct surrounding

98-percentile



50-percentile

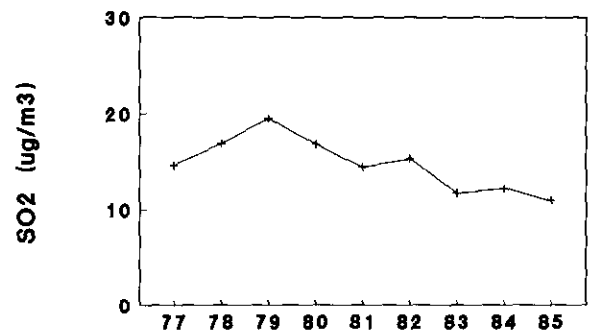


FIGURE 3. 50th and 98th percentile values of SO_2 ($\mu\text{g}/\text{m}^3$), 1977–1985.

Table 1. Yearly average concentrations^a and episodic concentrations of pollutants (2,3).

Component	Yearly, $\mu\text{g}/\text{m}^3$	Episodic, $\mu\text{g}/\text{m}^3$
O ₃	45	240-300
NO ₂	30	100-150
SO ₂	20	400-450
Sulfate	9	50-100
Nitrate	8	30- 50
Sulfuric acid	2	20- 50
Nitric acid	2	60- 70
Formic acid	1.5	12- 14
Acetic acid	1.5	12- 14
Propanoic acid	0.4	3- 4

^aSpatially averaged for the Netherlands.

areas (spatial resolution $15 \times 15 \text{ km}^2$), as well as on the scale of northwestern Europe (spatial resolution about $60 \times 60 \text{ km}^2$). The resulting concentration and deposition pattern can directly be compared with the results of the monitoring network.

A number of stations in the network are equipped with meteorological instruments (wind speed, wind direction, and global radiation). The availability of actual meteorological parameters enables us to run the transport models on-line. During episodes an up-to-date interpretation of the measured concentration fields in terms of source contributions can be given. Using prognostic meteorological information obtained from the Royal Netherlands Meteorological Institute (KNMI), an air pollution forecast can be given.

In the development of the models, an operational structure was a prerequisite. This means that only meteorological data that are available on a routine base are used as model input. This requirement hampers a more thorough description of several physical and chemical processes (e.g., the coarse vertical resolution, pseudolinear chemistry); however, in our opinion, this disadvantage is fully counterbalanced by the on-line applicability of the models.

Model Description

The long-range transport model (4) is based on a model developed by van Egmond and Kesseboom (5) and has been developed in cooperation with KNMI. The model covers an area of $2000 \times 2000 \text{ km}^2$. The output of the transport model consists of hourly averaged concentration fields of NO_x, NO₂, SO₂, and their oxidation products, NO₃ (sum of nitrate aerosol and nitric acid), and sulfate (SO₄). Dry and wet deposition fields of total sulfur and total oxidized nitrogen compounds are computed on a daily basis.

The vertical structure of the model (Fig. 4) consists of four layers. The surface layer accommodates modifications of the vertical profile at the surface due to dry deposition, for example. The mixing layer extends to the inversion height. During the night a constant mixing height is assumed. At daytime the mixing

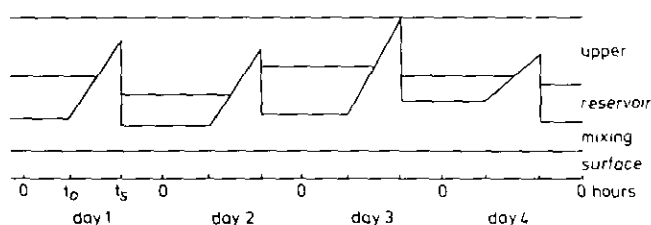


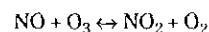
FIGURE 4. Schematic representation of the vertical structure of the transport models and the diurnal variations in mixing height.

height increases due to the incoming heat flux. Advection in the mixing-layer is described by the 1000 mbar windfield. Directly above the mixing layer, a reservoir layer is defined. In this layer the nighttime emissions of high point sources are stored. The depth of the reservoir layer is determined by the effective height of the point sources. The fourth and upper layer serve as a semipermanent reservoir for pollutants released from the mixing layer during the afternoon stratification. Transport in the reservoir and upper layer is described by the 850 mbar windfield.

Emission data are based on the Economic Commission for Europe/European Monitoring and Evaluation Program (ECE/EMEP) inventories (6), with exception of the Netherlands and its surroundings, where use was made of the more detailed TNO-emission inventory (7).

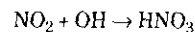
The transformation of SO₂ to sulfate is assumed to take place by three mechanisms: a) gas-phase oxidation by hydroxyl and organic radicals—the transformation rate is assumed to be proportional to the global radiation; b) oxidation at aerosol surfaces, catalyzed by transition metals, with a fixed rate of 0.5%/hr; c) oxidation in cloud water—this process is assumed to be operational in the upper layer only with a constant transformation rate of 1%/hr.

The concentrations of NO, NO₂, and O₃ are governed by the photostationary equilibrium:



The conservative components NO_x (sum of NO and NO₂) and Ox (sum of O₃ and NO₂) are independent of this equilibrium. From the modeled concentrations of NO_x and Ox and an effective equilibrium constant that is assumed to be proportional to the global radiation, the concentrations of NO and NO₂ are computed.

The formation of nitric acid is described by:



where the OH concentration is proportional to the global radiation. Nitrate formation by a reaction chain that is initiated by the reaction of NO₂ with ozone is also taken into account. In the latter oxidation process, a steady-state concentration of the intermediate radicals NO₃ and N₂O₅ is assumed.

Model Applications

The regional scale model has been applied to several winter and summer episodes. A fair agreement is found between observed and predicted SO_2 and SO_4 concentrations, both for the spatial distribution and for the temporal variation. The quality of NO_x predictions varies strongly: A reasonable agreement is found for summer episodes, but for winter episodes the model shows an underprediction of NO_x levels and an even larger underprediction of NO_2 levels, although the variations in time and space are in agreement with the available measurements. During winter episodes high NO_2 concentrations are frequently observed (8,9). These extremely high concentrations cannot be explained by the photostationary equilibrium using typical winter values for the background oxidant concentrations. It seems to be more likely that under these conditions the direct emission of NO_2 or the NO oxidation in the initial phase of the plume are enhanced (10,11). Effects like this are not included in the emission inventories. Additionally, the uncertainties in inversion height may account for the discrepancy found between observations and predictions. Further studies to improve the quality of the NO_x predictions are being undertaken.

As an example of the model results, the calculated SO_4 concentration fields for February 17, 1984, are shown in Figure 5 in combination with the observed concentrations (provided by ECE/EMEP). The time series for some European stations during the January 1985 episode are given in Figure 6.

Exposure Estimates

The results of the network have been used in longitudinal epidemiological studies on prevalence of respiratory symptoms and pulmonary function decline that have been carried out over a period of more than 15 years in two areas of the Netherlands with different levels of air pollution. These areas are the polluted area of Vlaardingen, a small city in the Rotterdam-Rijnmond area, and Vlagtwedde, a rural (residential) area in the northeast. Cohorts of about 2000 men and women were followed and examined for intervals of 3 years. Since the start of this study there has been a decrease in pollution levels in Vlaardingen. The significantly greater decline in FEV_1 (forced expiratory volume in 1 sec) in Vlaardingen in the first 9-year period could not be demonstrated in the last 6-year period (12). This is due mainly to lower FEV_1 values in the rural area in the last two investigations than might be expected on the basis of the first four surveys. Still, the FEV_1 values in the polluted area are consistently lower, which might point to long-term effects of the higher levels of air pollution in this area (12). An epidemiological study on the acute effects of air pollutants during episodes on pulmonary function of primary school children and adults has recently been started.

Both epidemiological studies and controlled exposure studies in humans and animals revealed that physiologic responses to sulfate aerosol are dependent upon aerosol acidity (13). In order to draw conclusions about response-exposure relationships, the population exposure to acidic aerosol has to be known with satisfactory accuracy. Exposures can be estimated from transport models or from measurements but direct measurement data on acidic aerosol are still not widely available. In case transport models are applied for exposure estimates, the following limitations have to be considered.

First, the model calculates concentrations that are representative for a larger area: The influence of local sources is only included in an averaged way. For a component such as NO_2 local sources (traffic) may contribute substantially to the local concentration and hence to the total exposure. However, for secondary pollutants such as sulfate and nitrate, the observed spatial gradients are weak and the direct influence of local sources will be small.

Second, in the prediction of personal exposures, an indoor/outdoor correction has to be applied according to individual activities. Model results with a time resolution of 1 hr are more suitable for this than the routine measurements with a time resolution of 24 hr. The sulfate concentrations may show large diurnal variations (14). When daily averaged concentrations are used for exposure estimates, large errors may easily be introduced. For example, on February 25, 1982, the model predicts a daily average sulfate concentration of $57.4 \mu\text{g}/\text{m}^3$ over the Netherlands (measured value $56.7 \mu\text{g}/\text{m}^3$). During the day, the modeled concentration covers a range of $25 \mu\text{g}/\text{m}^3$ (1:00 Central European Time) to $89 \mu\text{g}/\text{m}^3$ (16:00 Central European Time).

Furthermore, the acidic aerosol concentration is not directly calculated by the model; only total sulfate concentrations are modeled. An estimate of the sulfuric acid concentrations can be made by using an empirically derived degree of neutralization given by the molar ratio, $x = (\text{NH}_4 - \text{NO}_3 - \text{Cl})/\text{SO}_4$. The molar ratio x can take any value between 0 (pure sulfuric acid) and 2 (pure ammonium sulfate). However, taking into account the accuracy of the speciation measurements, it seems questionable to reach definitive conclusions regarding the acidity of the sulfate aerosol (14). Another approach is to model the sulfuric acid directly. To this end, concentrations of ammonia and ammonium have to be modeled also. Further development of the model in this sense is planned in the near future. Nevertheless, as it has been shown that sulfate has statistical significance as predictor of response to air pollution (15), it can be assumed that any relative trend found in sulfate concentrations under various emission scenarios will reflect the trend to be expected in population exposures.

Finally, only the inhalable fraction must be considered in exposure estimates to acidic aerosols. As secondary originated aerosol is mostly in the fine fraction ($< 2.5 \mu\text{m}$), this does not limit the applicability

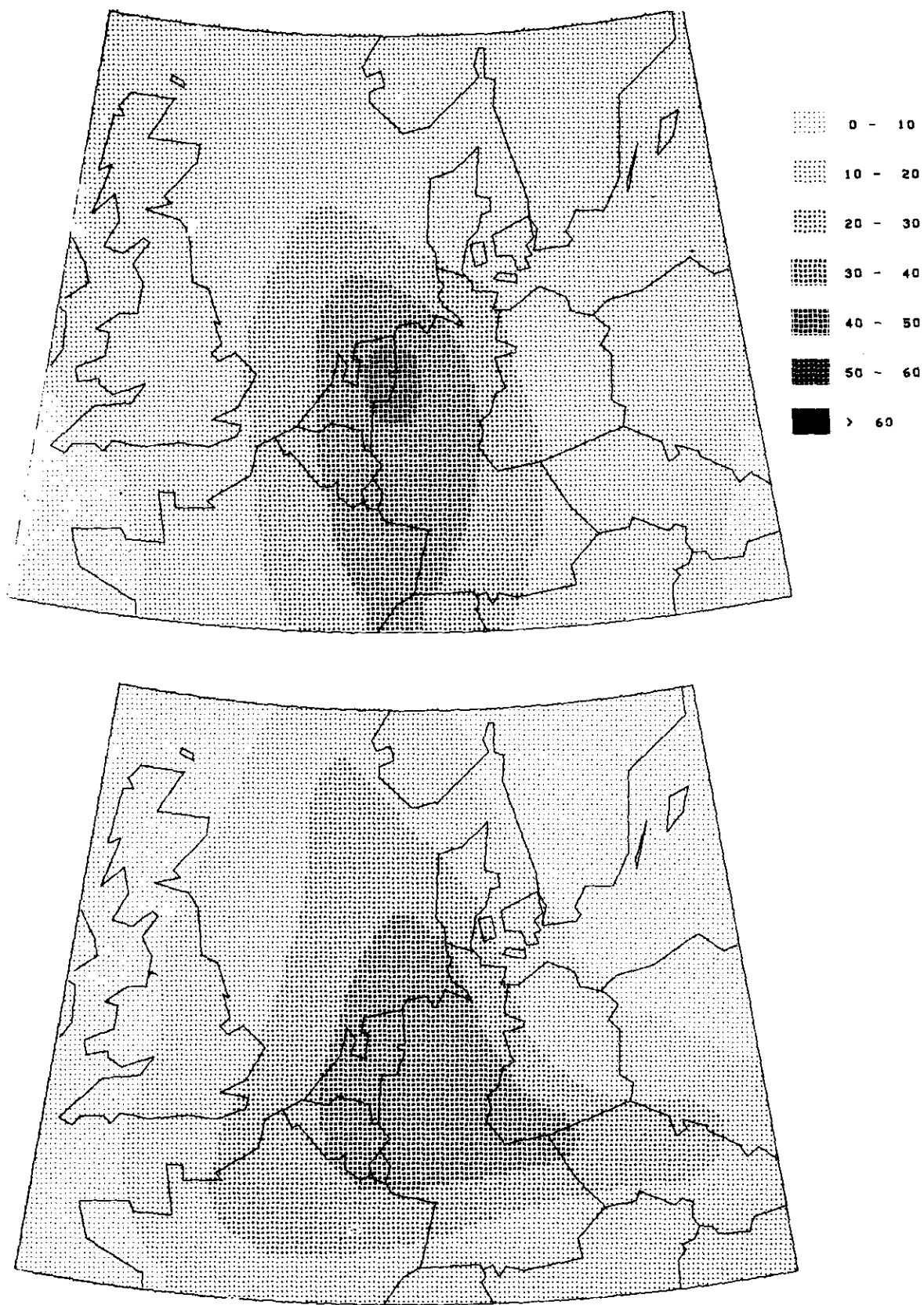


FIGURE 5. Observed and calculated SO_4 concentration fields (daily average, $\mu\text{g}/\text{m}^3$), February 17, 1984.

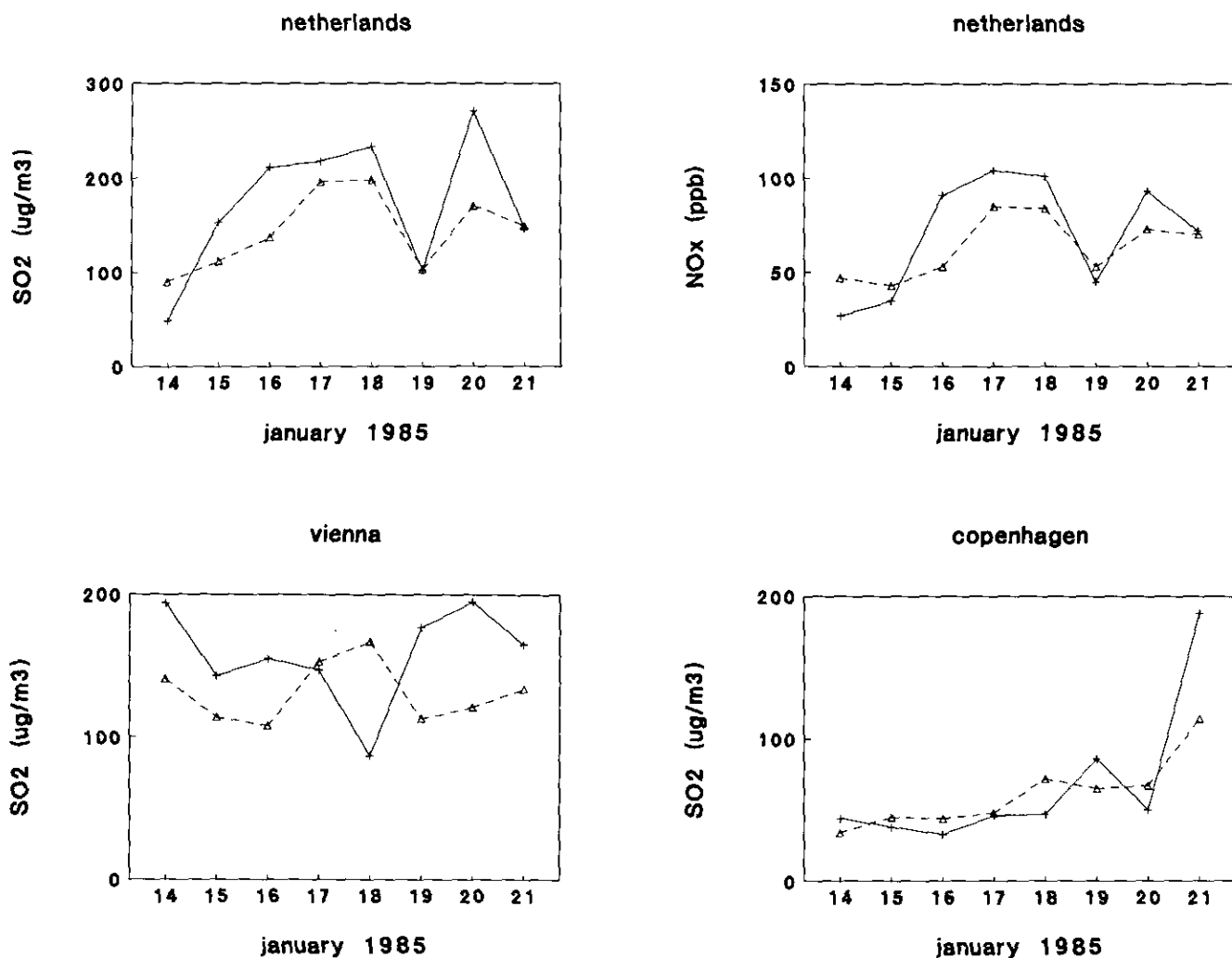


FIGURE 6. Measured (—) and calculated (---) concentrations during the period January 14–21, 1985.

of the model results in exposure estimates.

Notwithstanding the restrictions of model predictions previously described, these predictions, in combination with a limited number of (expensive) aerosol measurements, can be used to improve the knowledge of the temporal and spatial distribution of acidic aerosols and to estimate exposures to these aerosols.

REFERENCES

- van Egmond, N. D., and Onderdelinden, D. Objective analysis of air pollution monitoring network data; spatial interpolation and network density. *Atmos. Environ.* 15: 1035–1046 (1981).
- Air Quality Report 1984 and 1985. National Institute of Public Health and Environmental Protection (RIVM), Bilthoven, Netherlands. Report 228216052, 1986 (in Dutch).
- Diederer, H. S. M. A. Photochemical Air Pollution and Toxicity. Report CMP 84/31, Netherlands Organization for Applied Scientific Research, Delft, Netherlands, 1984 (in Dutch).
- de Leeuw, F. A. A. M. The Dutch aerosol study: Modeling aspects. In: *Aerosols: Research, Risk Assessment and Control Strategies* (S. D. Lee, T. Schneider, L. D. Grant, and P. J. Verkerk, Eds.), Lewis Publishers, Chelsea, 1986, pp. 301–315.
- van Egmond, N. D., and Kesseboom, H. Mesoscale air pollution dispersion models—I. Eulerian grid model. *Atmos. Environ.* 17: 257–265 (1983).
- Eliassen A., Hov, O., Isaksen, I. S. A., Saltbones, J., and Stordal, F. A lagrangian long-range transport model with atmospheric boundary layer chemistry. *J. Appl. Meteorol.* 21: 1645–1661 (1982).
- Huldy, H. J., and Veldt, C. Rekenstelsysteem luchtverontreiniging VI: Emissies. Report CMP 80/8, TNO, Delft, Netherlands (in Dutch).
- de Leeuw, F. A. A. M., and van Egmond, N. D. Air Pollution Episode 16–21 January 1985; Measurements, Model Calculations and Information Exchange. National Institute of Public Health and Environmental Protection (RIVM), Bilthoven, Netherlands. Report 228216042, 1985 (in Dutch).
- Onderdelinden, D., Blom, W. F., and van Rheineck Leyssius, H. J. Air Pollution Episode 14–24 January 1987: Concentrations of SO₂ and NO₂ in the Netherlands and Direct Surroundings. National Institute of Public Health and Environmental Protection (RIVM), Bilthoven, Netherlands. Report 228702001, 1987 (in Dutch).
- Lenner, M., Lindqvist, O., and Rosen, A. The NO₂/NO_x ratio in emission from gasoline powered cars: High NO₂ percentage in idle engine measurement. *Atmos. Environ.* 17: 1395–1398 (1983).
- Lindqvist, O., Ljungström, E., and Svensson, R. Low temperature thermal oxidation of nitric oxide in polluted air. *Atmos. Environ.* 16: 1957–1972 (1982).
- van der Lende, R., Schouten, J. P., Rijcken, B., and van der Meulen, A. Longitudinal epidemiological studies on effects of air pollution in the Netherlands. In: *Aerosols: Research, Risk*

- Assessment and Control Strategies (S. D. Lee, T. Schneider, L. D. Grant, and P. J. Verkerk, Eds.), Lewis Publishers, Chelsea, 1986, pp 731-742.
13. Lippmann, M. Airborne acidity: Estimates of exposure and human health effects. *Environ. Health Perspect.* 63: 63-70 (1985).
 14. van der Meulen, A. The Dutch aerosol study: Physical aspects. In: *Aerosols: Research, Risk Assessment and Control Strategies* (S. D. Lee, T. Schneider, L. D. Grant, and P. J. Verkerk, Eds.), Lewis Publishers, Chelsea, 1986, pp. 283-300.
 15. Ozkaynak, H., and Spengler, J. D. Analysis of health effects resulting from population exposures to acid precipitation precursors. *Environ. Health Perspect.* 63: 45-55 (1985).