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## The LOTOS–EUROS model: description, validation and latest developments

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**Abstract:** In this paper, we present a validation study for oxidants and secondary inorganic aerosols using the new regional Chemistry Transport Model (CTM) Long Term Ozone Simulation (LOTOS) European Operational Smog (EUROS). The model is the product of the integration of the Netherlands Organisation for Applied Scientific Research (TNO) LOTOS model and the National Institute for Public Health and the Environment (RIVM) EUROS model. In addition to a model description, we present an overview of a validation of the model. The new model is able to capture the variability of ozone well, especially in summer. However, the exchange between the mixing layer and the free troposphere was assessed to be too low resulting in too high ozone levels at days with low ozone. Sulphate levels are underestimated, whereas the concentrations of nitrate and ammonium are in line with measured data. The performance of LOTOS-EUROS is similar to other regional models in Europe. Examples of new applications of the model are presented. These include the modelling of sea salt and heavy metals.

**Keywords:** chemistry transport model; ozone; aerosol; validation.

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## 1 Introduction

The development and application of Chemistry Transport Models (CTMs) have a long tradition inside and outside Europe. Operational CTMs play an important role in the development of national and international policies to abate air pollution levels. For example, the EMEP model (Simpson et al., 2003) provides a scientific basis for the convention of Long-Range Transboundary Air Pollution (LRTAP). In the Netherlands, two operational models, Long Term Ozone Simulation (LOTOS) and European Operational Smog (EUROS) were independently developed by Netherlands Organisation for Applied Scientific Research (TNO) and National Institute for Public Health and the Environment (RIVM), respectively. Operational models are defined as models that contain in principle all relevant processes and that are capable of calculating a large number of scenarios on an hour-by-hour basis over extended periods of at least a year. Several operational models are currently available in Europe and have been compared for photo-oxidants and for aerosols (Hass et al., 2003; Roemer et al., 2003; van Loon et al.,

2004). Moreover, a regional scale model intercomparison to analyse the responses of different CTMs to emission changes/scenarios is ongoing in the EURO-DELTA program.

The LOTOS model has participated in these intercomparison studies. The LOTOS model, of which the basis has been created in the USA at Caltech in the 1970s of the last century (Reynolds et al., 1973), has been developed by TNO. The first operational version was available in 1988 and focused on ozone (Bultjes, 1992). Aerosols have been included since 1998/1999 (Schaap et al., 2004a,b). Data assimilation schemes, based on the Kalman Filter approach and focused on ozone, were integrated with the LOTOS model around 1999 (van Loon et al., 2000).

The EUROS model was developed by RIVM as a transport model for winter smog periods (de Leeuw and van Rheineck Leyssius, 1990). Later versions included ozone (van Loon, 1995), Persistent Organic Pollutants (POPs) (Jacobs and van Pul, 1996), aerosols (Matthijssen et al., 2002) and data-assimilation (Hanea et al., 2004).

Both LOTOS and EUROS cover Europe and extend to about 3.5 km in the vertical following the dynamic mixing layer approach. In addition, both models treat the chemistry with a modified version of the Carbon Bond Mechanism IV (CBMIV) (Whitten et al., 1980) in combination with a thermodynamic equilibrium module for semi-volatile aerosol species. Broadly, similar approaches are followed for dry and wet deposition and transport. Input on emissions and land use were almost identical. The largest differences were the projection, meteorological data and the technical structure of the models. Overall, the LOTOS and EUROS models have a lot in common. Moreover, the application area of the models is basically the same. On the basis of these facts, the already long cooperation on the working level led to a formal agreement between RIVM/MNP and TNO with the aim to integrate both models into one common LOTOS-EUROS model. In the new model LOTOS and EUROS are integrated incorporating modules from both models. In a few cases, both formulations have been kept, to be able to have the possibility to run both options. The philosophy of the LOTOS-EUROS model, like its similar models, is to have an operational model of intermediate complexity in the sense that the relevant processes are parameterised in such a way that the computational demands are moderate on a PC.

In the following sections, a description of the LOTOS-EUROS model will be given, followed by the results of a model validation exercise. This paper ends with results of a few recent developments and applications.

## 2 The LOTOS-EUROS modelling system

The LOTOS-EUROS modelling system can be applied for the following components:

- *Oxidants*: O<sub>3</sub>, VOCs, NO<sub>x</sub>, HNO<sub>3</sub>, etc.
- *Secondary Inorganic Aerosol (SIA)*: SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub>
- *Secondary Organic Aerosol (SOA) from terpenes*
- *Primary aerosol*: PM<sub>2.5</sub>, PM<sub>10-2.5</sub>, Black Carbon (BC), sea salt
- *Heavy metals*: Cd, Pb and other non-volatile metals
- *POPs*: BaP... etc.

Below we present an overview of the LOTOS–EUROS modelling system for these application areas except for POPs, which will be reported elsewhere. For an extensive description and details we refer to the documentation of the model (Schaap et al., 2005).

### 2.1 Continuity equation

The main prognostic equation in the LOTOS–EUROS model is the continuity equation that describes the change in time of the concentration of a component as a result of transport and diffusion, chemistry, dry and wet deposition, emissions and entrainment. In the model the continuity equation is solved by means of operator splitting. The time step is split in two parts, in which the concentrations changes are calculated for the first half and in reverse order for the second half.

### 2.2 Domain

The master domain of LOTOS–EUROS is bound at 35° and 70° North and 10° West and 60° East. The projection is normal longitude–latitude and the standard grid resolution is 0.50° longitude  $\times$  0.25° latitude, approximately 25  $\times$  25 km. By means of a control file the actual domain for a simulation can be set as long as it falls within the master domain as specified above. In addition, it is possible to increase or decrease the resolution up to a factor 8 or 2, respectively.

In the vertical, there are three dynamic layers and an optional surface layer. The model extends in vertical direction 3.5 km above sea level. The lowest dynamic layer is the mixing layer, followed by two reservoir layers. The height of the mixing layer is a part of the diagnostic meteorological input data. The height of the reservoir layers is determined by the difference between ceiling (3.5 km) and mixing layer height. Both layers are equally thick with a minimum of 50 m. In a few cases, when the mixing layer extends near or above 3500 m the top of the model exceeds the 3500 m according to the above-mentioned description. Optionally, a surface layer with a fixed depth of 25 m can be included in the model. Simulations with the surface layer have 4 layers (z4), otherwise 3 layers (z3).

For output purposes, the concentrations at measuring height (reference height is usually 3.6 m) are diagnosed by the constant flux approach that relates the dry deposition speed and the concentration of a pollutant.

### 2.3 Processes

#### 2.3.1 Transport

The transport consists of advection in three dimensions, horizontal and vertical diffusion and entrainment. The advection is driven by meteorological fields that are input every 3 hr. The vertical wind speed is calculated by the model as a result of the divergence/convergence of the horizontal wind fields. The recently improved and highly accurate, monotonic advection scheme developed by Walcek (2000) is used to solve the system. The number of steps in the advection scheme is chosen such that the Courant restriction is fulfilled.

Entrainment is determined by the growth of the mixing layer during the day. Each hour the vertical structure of the model is adjusted to the new mixing layer depth. After the new structure is set the pollutant concentrations are redistributed using linear interpolation.

The horizontal diffusion is described with a horizontal eddy diffusion coefficient following the approach by Liu and Durran (1977). Vertical diffusion is described using the standard  $K_z$ -theory. The  $K_z$  values are calculated in the stability parameterisation (see below). Vertical exchange is calculated employing the new integral scheme by Yamartino et al. (2004).

### 2.3.2 *Chemistry*

The LOTOS–EUROS model contains two chemical mechanisms, the TNO CBM-IV scheme (Schaap et al., 2005) and the CBM-IV by Adelman (1999). In this study, we used the TNO CBM-IV scheme that is a modified version of the original CBM-IV (Whitten et al., 1980). The scheme includes 28 species and 66 reactions, including 12 photolytic reactions. Compared to the original scheme, steady-state approximations were used to reduce the number of reactions. In addition, reaction rates have been updated regularly. The mechanism was tested against the results of an intercomparison presented by Poppe et al. (1996) and found to be in good agreement with the results presented for the other mechanisms. Aerosol chemistry is represented using ISORROPIA (Nenes et al., 1999) and SORGAM (Schell et al., 2001).

### 2.3.3 *Dry and wet deposition*

The dry deposition in LOTOS–EUROS is parameterised following the well-known resistance approach. The deposition velocity is described as the reciprocal sum of three resistances: the aerodynamic resistance, the quasi-laminar layer resistance and the surface resistance. The aerodynamic resistance is dependent on atmospheric stability. The relevant stability parameters are calculated using standard similarity theory profiles. The quasi-laminar layer resistance and the surface resistances for acidifying components and particles are described following the EDACS system (Erisman et al., 1994).

Below cloud scavenging is described using simple scavenging coefficients for gases (Schaap et al., 2004a,b) and following Simpson et al. (2003) for particles. In-cloud scavenging is neglected due to the limited information on clouds. Neglecting in-cloud scavenging results in too low wet deposition fluxes but has a very limited influence on ground level concentrations (see Schaap et al., 2004b).

## 2.4 *Input data*

### 2.4.1 *Meteorological data*

The LOTOS–EUROS system is presently driven by 3-hr meteorological data. These include 3D fields for wind direction, wind speed, temperature, humidity and density, substantiated by 2D-gridded fields of mixing layer height, precipitation rates, cloud cover, several boundary layer and surface variables. The standard meteorological data are

produced in the Free University of Berlin employing a diagnostic meteorological analysis system based on an optimum interpolation procedure on isentropic surfaces. The system utilises all available synoptic surface and upper air data (Kerschbaumer and Reimer, 2003). Also, meteorological data obtained from ECMWF can be used to force the model.

#### 2.4.2 Emissions

The anthropogenic emissions used in this study are a combination of the TNO emission database (Visschedijk and Denier van der Gon, 2005) and the CAFE baseline emissions for 2000. For each source category (Snap 1) and each country, we have scaled the country totals of the TNO emission database to those of the CAFE baseline emissions. Hence, we use the official emission totals as used in the LRTAP protocol but we benefit from the higher resolution of the TNO emission database ( $0.25^\circ \times 0.125^\circ$  longitude–latitude). The total of annual emission is broken down to hourly emission, estimates using time factors for the emission's strength variation over the months, days of the week and the hours of the day (Buitjes et al., 2003).

In LOTOS–EUROS biogenic isoprene emissions are calculated following Veldt (1991) using the actual meteorological data. In addition, sea salt emissions are parameterised following Monahan et al. (1986) from the wind speed at 10 metres.

#### 2.4.3 Land use

The land use data used in LOTOS–EUROS are derived from the  $1.1 \times 1.1\text{-km}^2$  resolution land use database PELINDA (de Boer et al., 2000) and the IIASA database for Russia (Stolbovoi and McCallum, 2002). From these databases, the surface fraction covered by the land use classes used in the dry deposition module has been calculated for each cell in the domain.

#### 2.5 Run specification

To assess the performance of the LOTOS–EUROS model (version 1.1) the model was used to simulate the concentrations of oxidants and aerosols for the whole year of 1999. A summary of the validation for oxidants and SIA is given below. These components were chosen as measurements are easily accessible and the infrastructure for the comparison with data was already there from a recent model intercomparison study (van Loon et al., 2004). Furthermore, we present the results and a limited validation for sea salt and heavy metals.

### 3 Results

In this paper, we present the results of the LOTOS–EUROS system for oxidants. The modelled oxidant concentrations are compared to available data from the EMEP network. The consequences of the inclusion of the surface layer are presented and discussed.

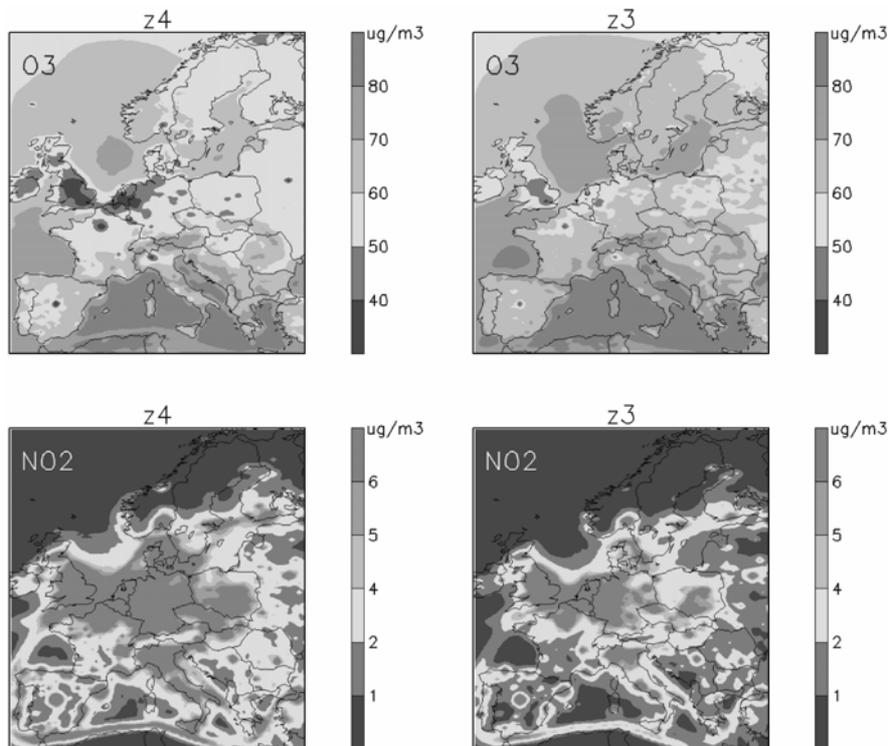
### 3.1 Oxidants

#### 3.1.1 Distributions

**3.1.1.1 Ozone:** Figure 1 shows the mean modelled  $O_3$  fields for the year 1999 for both the 4- and 3-layer versions of LOTOS-EUROS. Using the surface layer (z4) generally yields lower ozone concentrations without the surface layer. Most of the remote areas the lower modelled concentrations are due to the smaller reservoir (depth) that causes a faster decline in concentrations due to dry deposition. In the source areas, ozone is also strongly affected by the incorporation of a surface layer. Here, the ozone titration by NO is stronger as the volume, in which the emitted  $NO_x$  is mixed into, is smaller. This causes reductions in the mean ozone values of about 12%.

**3.1.1.2 Nitrogen oxides:** The modelled mean  $NO_2$  values (Figure 1) for the model with the added surface layer are higher than without this extra layer. The differences for  $NO_2$  plus NO can be as much as a factor 2. This can be explained by the smaller extent of the layer where the emissions of NO are mixed. Thus, the emissions of NO in the small surface layer are more concentrated than in the mixed boundary layer of the 3-layer version. As a consequence,  $NO_2$  formed from the reaction of NO with  $O_3$ , shows higher values in the four-layer version.

**Figure 1** Annual average distribution of ozone and  $NO_2$  for 1999 modelled with (z4) and without (z3) a surface layer



### 3.1.2 Validation

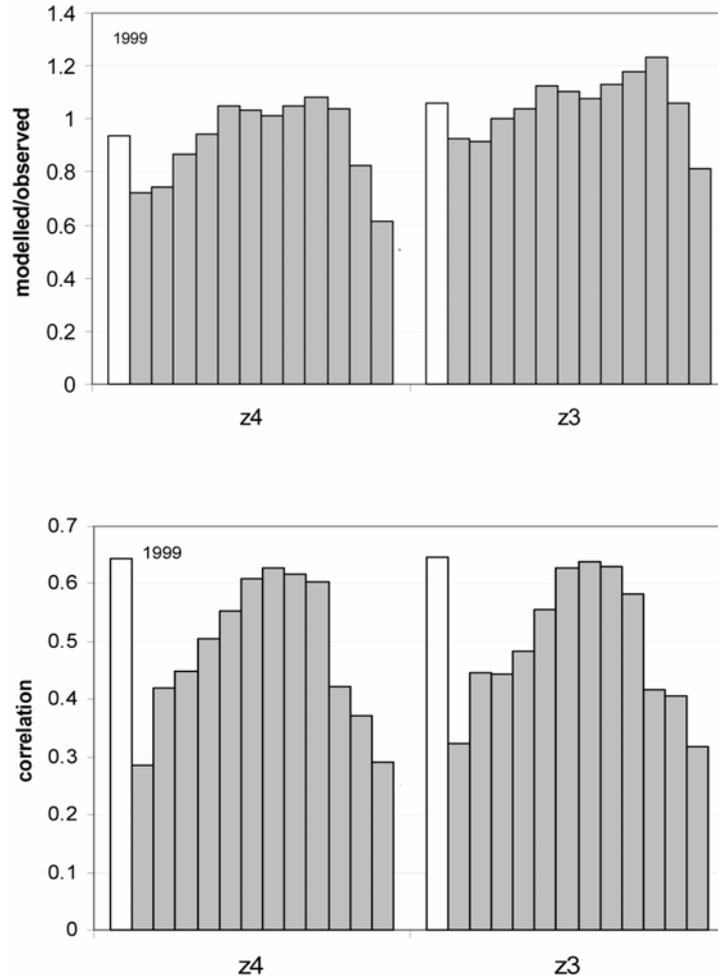
Below we compare the modelled concentrations of ozone, daily maximum ozone and NO<sub>2</sub> with observations obtained from the EMEP ([www.emep.int](http://www.emep.int)) and AIRBASE databases. The data selection and evaluation follows van Loon et al. (2004). We evaluate the modelled and measured mean concentrations. Further, the agreement of the model results with EMEP observations is represented by the Root Mean Square Error (RMSE) and the (temporal) correlation coefficient.

**3.1.2.1 Ozone:** In Table 1, we summarise the validation results for O<sub>3</sub> and daily maximum ozone concentrations. The modelled annual average ozone concentration is close to the observed value. On an average the results with four layers slightly underestimate annual mean ozone levels, whereas the three layer results slightly overestimate. Inspection of the performance with respect to mean concentrations as a function of season reveals a pattern with good agreement in summer, and an underestimation of the measured concentrations in winter (see Figure 2). This pattern is most pronounced for the simulation with the surface layer. In general, lower night time concentrations due to the inclusion of the surface layer cause a better overall agreement in summer, but also a higher underestimation in winter due to higher titration and a steeper gradient due to deposition. Correlation coefficients are very similar for the two simulations and tend to increase significantly for the day time and daily maximum values compared to the data for the whole day. For ozone, it is difficult to point out if the inclusion of a surface layer increases the performance significantly.

**Table 1** Statistical comparison between modelled and observed ozone and NO<sub>2</sub> concentrations for the model (LE) with (z4) and without (z3) a surface layer

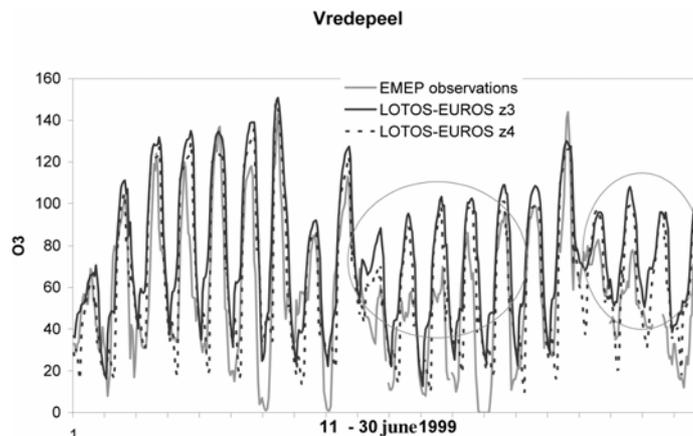
	<i>Observed mean</i>	<i>Modelled mean</i>	<i>RMSE</i>	<i>Correlation</i>
Whole day O <sub>3</sub>				
LE_z4	60.17	56.20	25.22	0.65
LE_z3	60.17	63.95	24.51	0.65
Daytime O <sub>3</sub>				
LE_z4	67.59	66.33	23.46	0.69
LE_z3	67.59	74.28	23.14	0.70
Day max O <sub>3</sub>				
LE_z4	81.92	78.88	20.39	0.75
LE_z3	81.92	82.91	19.92	0.75
Whole day NO <sub>2</sub>				
LE_z4	13.70	10.79	11.44	0.40
LE_z3	13.70	6.45	12.44	0.37

**Figure 2** Average modelled to measured mass ratio (upper panel) and correlation coefficient (lower panel) for ozone. The white bar represents the annual average whereas the grey bars indicate the seasonal variation (January–December)



A detailed analysis of the time series learns that the model is not able to reproduce the highest peak values. On the other hand, the model over-predicts the ozone peak values in low ozone periods in summer as indicated in Figure 3. These periods coincide with periods of cloudy and rainy weather. The model concentrations are much higher than the large scale ozone concentrations in the free troposphere (Logan, 1999) whereas the measured concentrations are quite closer to these values. Hence, this indicates that in these conditions the model underestimates the exchange between the free troposphere and the mixing layer. This could also explain a part of the underestimation in winter when ozone levels in the free troposphere are higher than calculated near the surface for the rural stations used in this study. It was observed in another study (City-Delta, Cuvelier et al., 2004) that the use of a surface layer improves modelling of ozone in high  $\text{NO}_x$  situations associated with large agglomerations.

**Figure 3** Comparison of the modelled and measured time series of ozone at Vredepeel, The Netherlands



**3.1.2.2 Nitrogen oxides:** The model systematically underestimates the concentrations of nitrogen oxides. The measured concentrations for  $\text{NO}_2$  are underestimated by 21% and 53% for the 4- and 3-layer versions, respectively. Hence, the inclusion of the surface layer improves the representation of ground level  $\text{NO}_2$  concentrations. Underestimation of  $\text{NO}_2$  concentrations is higher in summer than in winter. However, the low correlation coefficients ( $\sim 0.4$ ) between the model and measurements, that are in line with those of other models (van Loon et al., 2004), indicate that the modelling of  $\text{NO}_2$  with a regional model such as LOTOS–EUROS is still problematic. Inclusion of a surface layer only slightly increases the correlation coefficient that indicates that the vertical resolution of a model may not be the crucial factor to correctly model the temporal variability in  $\text{NO}_2$  concentrations.

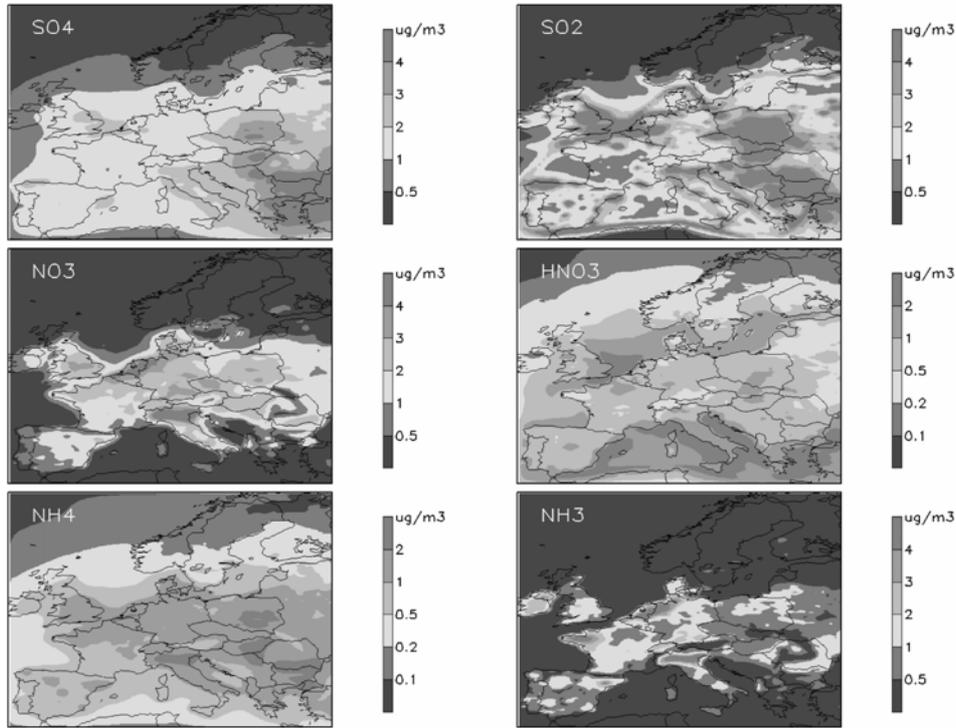
## 3.2 Secondary inorganic aerosol

The results of the LOTOS–EUROS system for SIA, are presented here. The modelled concentrations of sulphate, nitrate, ammonium and their precursors are compared to available data from the EMEP network. Further, sensitivity calculations are presented and discussed.

### 3.2.1 Distributions

Figure 4 shows the mean modelled SIA distributions for the year 1999 for the 4-layer version of LOTOS–EUROS. In addition, the precursor gas distributions are presented and described below.

**3.2.1.1 Sulphate:** The modelled sulphate concentrations are high in south-east Europe. Here, modelled concentrations exceed  $3 \mu\text{g}/\text{m}^3$  on average. Secondary maxima can be observed in northern Spain, central England, the Po-valley and the English Channel. More than a large amount of sulphate levels between  $1$  and  $2 \mu\text{g}/\text{m}^3$  are calculated in most of the places in Europe.

**Figure 4** Annual average distributions of SIA and their precursors for 1999

**3.2.1.2 Nitrate:** Maximum nitrate concentrations are found in the areas such as the Netherlands, Belgium, north western Germany, northern Italy, England, southern Germany and the Czech Republic, where the annual averaged concentrations exceed  $3\text{--}4\ \mu\text{g}/\text{m}^3$ . These areas are characterised by high ammonia emissions. Over central Europe, concentrations are typically between  $2$  and  $3\ \mu\text{g}/\text{m}^3$ . Over southern Europe, nitrate concentrations do not exceed  $2\ \mu\text{g}/\text{m}^3$ , except for northern Italy and a region over former Yugoslavia. Over most of Scandinavia, annual average nitrate concentrations are calculated to be lower than  $0.5\ \mu\text{g}/\text{m}^3$ , due to the low amount of nitric acid formed there. Nitrate is a continental phenomenon, as its concentration rapidly trails from coast to open sea.

**3.2.1.3 Ammonium:** Ammonium neutralises both sulphate and nitrate. Hence, the gradients in the distribution show similarities to those of the anions. Modelled ammonium concentrations are about  $2\ \mu\text{g}/\text{m}^3$  over most of Europe and trail towards remote locations such as the Open Ocean and Scandinavia. Ammonium peaks in those areas where nitrate and sulphate peak, that is, south-east Europe, the Po-Valley and the Benelux.

**3.2.1.4 Precursor gases:** Sulphur dioxide concentrations show high gradients over Europe. The distribution is characterised by concentrations above  $5\ \mu\text{g}/\text{m}^3$  near sources. These include large regions in (south-) Eastern Europe, shipping tracks, large cities and

heavily industrialised areas. Scandinavia and southern France are rather clean with levels less than  $1 \mu\text{g}/\text{m}^3$ . The sulphur dioxide distribution does not show a significant seasonal behaviour.

In continental regions the annual average nitric acid concentrations are mainly between  $0.5$  and  $1.0 \mu\text{g}/\text{m}^3$ , see Figure 4. Over the sea, the concentrations are much higher than those over land. Especially the ship tracks, in which large amounts of  $\text{NO}_x$  are emitted, are visible in the calculated nitric acid fields. In reality nitric acid in these regions may be associated with sea salt. In winter, computed concentrations of nitric acid are much lower than those of nitrate. In summer, only in north-west Europe nitric acid concentrations are well below those of nitrate. In other areas they are higher or comparable to those of aerosol nitrate.

Owing to the short atmospheric lifetime of ammonia, its concentration field strongly resembles its emission distribution (Figure 4). Maximum concentrations occur in the areas with the highest emissions, such as the Netherlands, southern England and the Po-valley. Only in these areas, annual mean concentrations of more than  $2 \mu\text{g}/\text{m}^3$  are calculated. Outside the source areas, the ammonia concentrations decline rapidly.

### 3.2.2 Validation

The model results of LOTOS–EUROS are compared to concentrations observed at regional background stations. Observations of SIA and  $\text{SO}_2$  used in this study are gathered from the EMEP database ([www.emep.int](http://www.emep.int)). Mountain stations were excluded from the analysis. The data for nitrate and ammonium were screened as the measurements of these compounds are prone to artefacts (Schaap et al., 2002, 2004a). Nitrate data obtained with cellulose filters were interpreted as total nitrate. Aerosol nitrate data from inert filters were used in this study, although we are aware of that they are prone to losses at temperatures above  $20^\circ\text{C}$ . Total nitrate and ammonium data that were not obtained in a single measurement set-up were disregarded. As the spatial coverage for aerosol nitrate and ammonium is low, all stations were included in the analysis. Hence, results for these components are biased to a few stations. A summary of the validation results is given in Table 2.

**3.2.2.1 Sulphate and sulphur dioxide:** The annual average calculated concentrations of sulphate and sulphur dioxide in LOTOS–EUROS is too low. The average correlation between measured and modelled concentrations is  $0.5$  for sulphate and  $0.4$  for sulphur dioxide. For  $\text{SO}_2$ , the model overestimates the observed concentrations at low  $\text{SO}_2$  concentrations, whereas at high  $\text{SO}_2$  concentrations, the model underestimates the observed concentrations (Figure 5). The underestimation of the measured sulphate concentrations is  $40\%$  on average (Figure 5) and peaks in Germany and especially in Spain. This underestimation is probably due to a much slower oxidation rate of  $\text{SO}_2$ . Therefore, the use of an explicit oxidation scheme of  $\text{SO}_2$  by  $\text{H}_2\text{O}_2$  and  $\text{O}_3$  are currently under research. On the other hand, in Spain, where the underestimation of sulphate is most severe, the measurements show that ammonium levels are much lower than one would expect, when ammonium sulphate was present. Hence, other sources and mechanisms for sulphate formation may be important there.

**Table 2** Statistical comparison between annual average modelled and observed SIA, SO<sub>2</sub>, total ammonium (TNH<sub>4</sub>) and total nitrate concentrations (TNO<sub>3</sub>) for 1999 for the model (LE) with (z4) and without (z3) a surface layer

	<i>Observed mean</i>	<i>Modelled mean</i>	<i>RMSE</i>	<i>Correlation</i>
SO <sub>4</sub>				
LE_z4	2.47	1.47	2.16	0.51
LE_z3	2.47	1.49	2.16	0.51
NH <sub>4</sub>				
LE_z4	1.15	1.16	1.00	0.44
LE_z3	1.15	1.20	1.00	0.44
NO <sub>3</sub>				
LE_z4	2.99	2.97	2.42	0.52
LE_z3	2.99	3.11	2.46	0.49
SO <sub>2</sub>				
LE_z4	2.71	2.32	3.36	0.40
LE_z3	2.71	1.94	3.23	0.41
TNH <sub>4</sub>				
LE_z4	1.65	1.29	1.93	0.39
LE_z3	1.65	1.10	1.93	0.41
TNO <sub>3</sub>				
LE_z4	1.83	1.72	1.69	0.46
LE_z3	1.83	1.76	1.70	0.47

The performance of the model for SO<sub>4</sub> does not change significantly by the inclusion of an extra surface layer. On the other hand for SO<sub>2</sub>, the model calculations increase by 16% when using the surface layer compared to the simulation without a surface layer. This is explained by the primary nature of SO<sub>2</sub>.

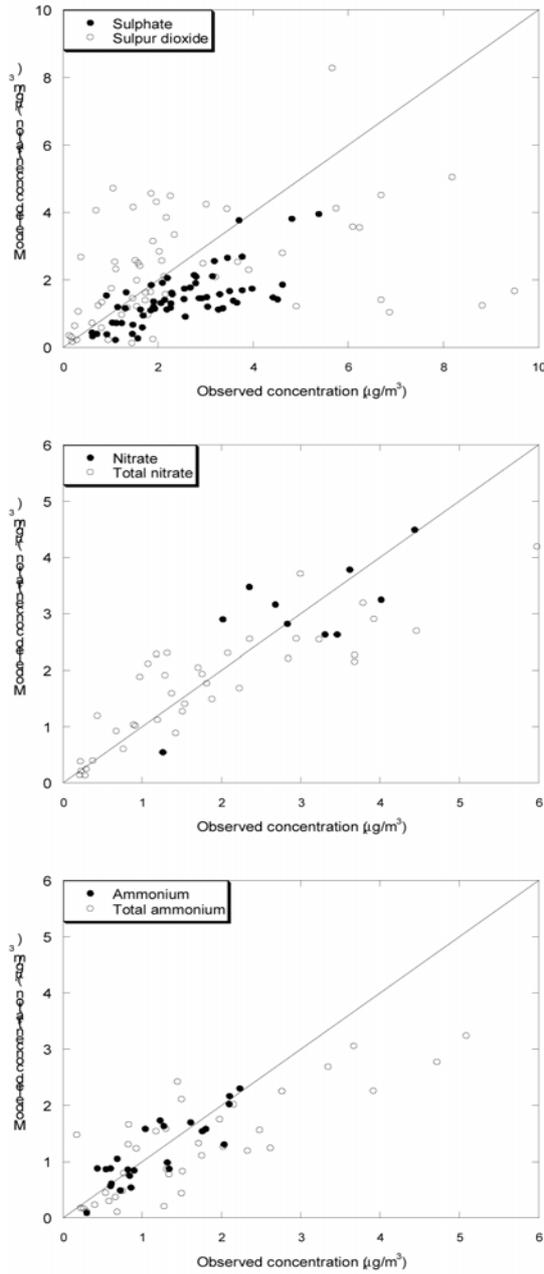
**3.2.2.2 (Total) Nitrate:** The annual average modelled NO<sub>3</sub> and total nitrate (TNO<sub>3</sub> = NO<sub>3</sub> + HNO<sub>3</sub>) concentrations are close to the observed values and can be seen in Table 2. The scatter plot (Figure 5) shows that the individual stations are well spread around the one-to-one line. The average correlation coefficients for total nitrate and NO<sub>3</sub> are around 0.5 indicating that the temporal variation is reasonably represented by the model.

As with sulphate, the performance of LOTOS–EUROS for nitrate is similar using 4 or 3 layers.

**3.2.2.3 (Total) Ammonium:** Table 2 and Figure 5 show that the annual average modelled concentrations of NH<sub>4</sub> are also closer to the observed values, whereas the modelled total ammonium concentrations underestimate the observed ones by 22% and 33% using 4 and 3 layers, respectively. Owing to the primary nature of NH<sub>3</sub>, the inclusion of a surface layer increases NH<sub>3</sub> and consequently total ammonium concentrations. The underestimation at the high end of the observations may also be influenced by emissions near the measurement stations. The average correlation for ammonium is 0.44 and for total ammonium around 0.4.

In general, as for  $O_3$  and  $NO_2$ , also for the inorganic aerosols, the performance of LOTOS–EUROS is comparable to the other CTMs who participated in the EMEP intercomparison study (van Loon et al., 2004).

**Figure 5** Comparison between annual average concentrations from observations and LOTOS–EUROS for SIA and precursors. Each dot represents a different station

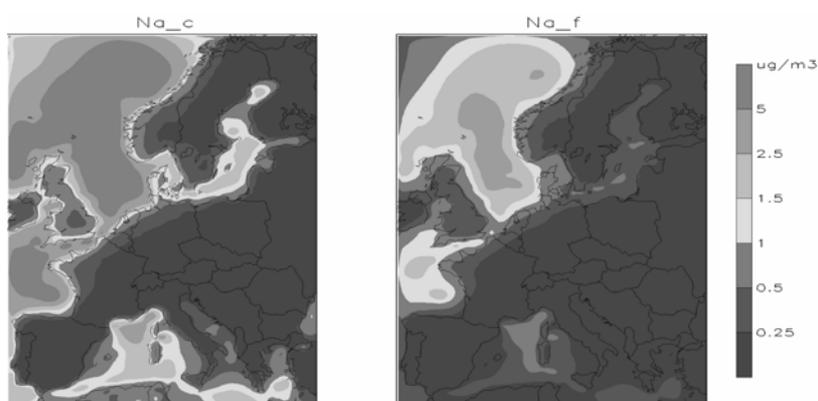


## 4 Recent applications

### 4.1 Sea salt

The primary aerosol distributions modelled by LOTOS–EUROS also encompass the sea salt distribution, that is represented by sodium ( $\text{Na}^+$ ) concentrations. A separation is made between coarse ( $>2.5 \mu\text{m}$ ) and fine ( $<2.5 \mu\text{m}$ ) mode  $\text{Na}^+$ . Figure 6 shows the yearly averaged fields concentrations for coarse and fine  $\text{Na}^+$  for 2001 from the LOTOS–EUROS model. The modelled annual average sodium concentrations (2001) are high over the open ocean but drop off fast over land.

**Figure 6** Distributions of coarse and fine  $\text{Na}^+$  for 2001 (in  $\mu\text{g}/\text{m}^3$ )



The modelled total (coarse+fine)  $\text{Na}^+$  concentrations are compared to the corresponding daily measurements from the three Danish EMEP stations and the five Norwegian EMEP stations in Table 3. For all stations, modelled values are higher than those observed. The difference between model and observations is larger for the Norwegian stations than for the Danish stations. The correlation coefficients are mostly well above 0.5. In Figure 7 modelled and observed  $\text{Na}^+$  concentrations at the Danish station Tange are plotted for the year 2001. The variability in the observed concentrations is well captured by the model that is reflected in a high correlation coefficient of 0.67. However, most observed peak values are overestimated by the model causing the positive bias between LOTOS–EUROS and the observations.

We have also compared our model results to a few observations that cover different periods than the model results (see Table 3). These data confirm the findings for the EMEP sites.

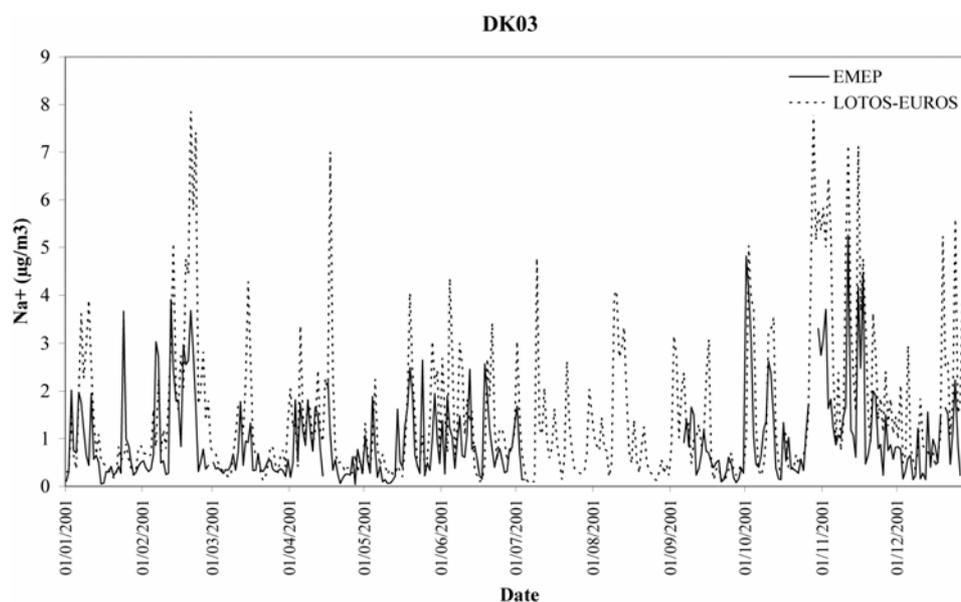
Our results are comparable to the results from a comparison study by Foltescu et al. (2005) between the MATCH model and EMEP observations. The over estimation of the peak concentrations may be related to the relatively large uncertainty associated with the wind speed dependent emission strength of sea salt particles. Further, our model assumes a coarse mode size distribution that may not be representative for the size distribution directly after emission that may yield an underestimation of the deposition (of the largest particles) in the source regions. The orography around the Norwegian stations may largely explain the less favourable comparison between modelled and measured concentrations than in Denmark.

The results shown here only comprise a few stations. To allow a better evaluation of the model, a more extensive validation, including the literature study is foreseen in the near future.

**Table 3** Comparison between average observed and modelled  $\text{Na}^+$  concentrations ( $\mu\text{g}/\text{m}^3$ ) from both observations and model and correlation between the modelled and observed daily concentrations. All the data represent 2001 unless stated otherwise and have a data completeness of 80% or more

EMEP Stations		Observed mean	Modelled mean	Correlation
DK03		0.96	1.50	0.67
DK05		1.44	1.64	0.29
DK08		1.40	2.36	0.66
NO01		0.33	1.08	0.57
NO08		0.32	1.57	0.65
NO15		0.26	1.25	0.56
NO39		0.15	1.16	0.51
NO41		0.09	0.37	0.21
Extra stations	Period of observation	Observed mean	Modelled mean	Reference for observations
Cabauw (NL)	Winter 2000	0.81	1.00	Weijers et al., 2002
Speuld (NL)	1995	0.79	1.11	Erisman et al., 1996
Duisburg (D)	2002–2003	0.66	0.50	Quass et al., 2004

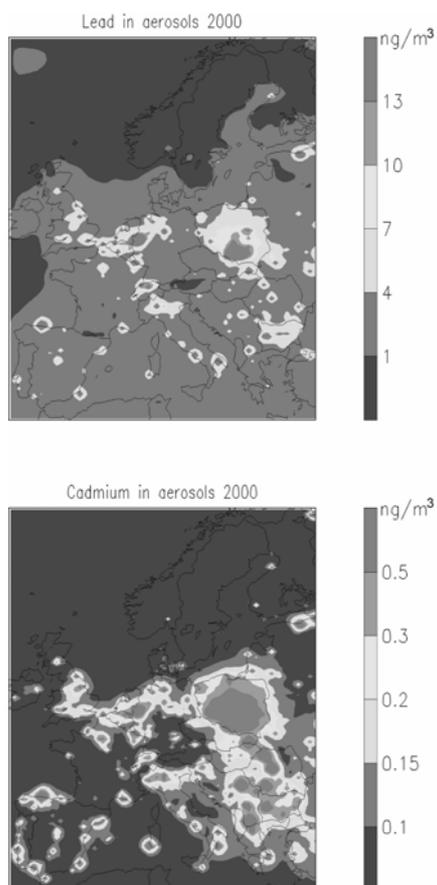
**Figure 7** Comparison of modelled and observed  $\text{Na}^+$  concentrations at EMEP station Tange (DK03) in Denmark. The solid line indicates the observations. The dashed line indicates the modelled values by LOTOS-EUROS



#### 4.2 Heavy metals

The distribution of lead and cadmium is also calculated by the LOTOS–EUROS model. Anthropogenic emissions are derived from the TNO/EMEP emission inventory (Denier van der Gon et al., 2005). Natural emissions and boundary conditions are set according to Travnikov and Ilyin (2005). Figure 8 shows the calculated annual mean distributions for lead and cadmium for the year 2000.

**Figure 8** Modelled concentrations of lead (top) and cadmium (bottom) averaged over the year 2000 in  $\text{ng}/\text{m}^3$



A comparison with measurements performed at several EMEP stations revealed that the model values for both cadmium and lead are well below the measured concentrations. The underestimation is on average a factor 3.3 and reaches up to a factor 8 for a few specific sites. This discrepancy has also been observed between other models and observations (Ilyin and Travnikov, 2005) and is believed to be largely due to the high-level of uncertainty of emission data. The origins of the discrepancies will be further researched with special attention to the uncertainties in the emission estimates. Moreover, optimising the emissions estimates such that the modelling results are in line with observed concentrations will be considered.

## 5 Conclusions and outlook

In this paper, we have presented the new LOTOS–EUROS model that is the product of the integration of the LOTOS and EUROS models. From the initial results we have concluded that:

- The variability in ozone concentrations is well reproduced by the model. Ozone concentrations are overestimated at days with low ozone that is probably associated with an underestimation of vertical mixing in the presence of clouds.
- The use of a surface layer results in a better representation of primary emitted species.
- Overall, for SIA, the agreement between modelled and observed concentrations is reasonable. Sulphate concentrations are underestimated by the model whereas nitrate and ammonium are in agreement with observations. We have observed that sulphate and ammonium concentration data do not support the model hypothesis that sulphate is present as ammonium sulphate in Spain explaining the better comparison for ammonium.
- The performance of LOTOS–EUROS is similar to other regional models in Europe.
- The first simulations for sea salt show promising results. The timing of the events is good but concentrations are overestimated.
- Modelled heavy metal concentrations underestimate measured values by a factor 3–4 that reflects the current status of emission estimates.

### 5.1 Outlook

The new model will be used for policy support and research applications at both TNO and RIVM. The model results of a new model will be compared to other models in the EURODELTA exercise. Further (planned) applications and investigations are:

- to improve the mixing description due to convective clouds and investigate the sensitivity to several sulphate formation mechanisms
- to operationally forecast air pollution levels over Europe ( $O_3$ ,  $NO_2$  and PM)
- to couple the model with the integrated assessment model RAINS-NL
- to integrate in situ and satellite observations and the model into one system using data assimilation techniques. The system will be used to improve the distributions of  $PM_{2.5}$ ,  $O_3$  and  $NO_2$  over Europe
- to include aerosol dynamics in the model description.

## References

- Adelman, Z.E. (1999) 'A re-evaluation of the Carbon Bond-IV photochemical mechanism', MSc Thesis, Department of Environmental Sciences and Engineering, School of Public Health, University of North Carolina, USA.
- Builtjes, P.J.H. (1992) 'The LOTOS-Long Term Ozone Simulation-project Summary Report', *TNO Report TNO-MW-R92/240*.

- Builtjes, P.J.H., van Loon, M., Schaap, M., Teeuwisse, S., Visschedijk, A.J.H. and Bloos, J.P. (2003) 'Project on the modelling and verification of ozone reduction strategies: contribution of TNO-MEP', *TNO-report, MEP-R2003/166*, Apeldoorn, The Netherlands.
- Cuvelier, C., Thunis, P., Stern, R., Moussiopoulos, N., Builtjes, P., Rouil, L., Bedogni, M., Tarrason, L., Amann, M. and Heyes, C. (2004) 'City-delta phase 2', *Draft Final Report*, IIASA Contract No. 03-137.
- de Boer, M., De Vente, J., Múcher, C.A., Nijenhuis, W. and Thunnissen, H.A.M. (2000) 'Land cover monitoring. An approach towards pan European land cover classification and change detection', *NRSP-2, Project 4.2/DE-03*.
- de Leeuw, F.A.A.M. and van Rheineck Leyssius, H.J. (1990) 'Modeling study of SO<sub>x</sub> and NO<sub>x</sub> during the January 1985 smog episode', *Water, Air and Soil Pollution*, Vol. 51, pp.357–371.
- Denier van der Gon, H.A.C., van het Bolscher, M., Visschedijk, A.J.H. and Zandveld, P.Y.J. (2005) 'Study to the effectiveness of the UNECE heavy metals protocol and costs of possible additional measures, phase 1: estimation of emission reduction resulting from the implementation of the HM protocol', *TNO Report B&O-A R 2005/193*.
- Erisman, J.W., Draaijers, G.P.J., Mennen, M.G., Hogenkamp, J.E.M., van Putten, E., Uiterwijk, W., Kemkers, E., Wiese, H., Duyzer, J.H., Otjes, R. and Wyers, G.P. (1996) 'Towards development of a deposition monitoring network for air pollution in Europe', *RIVM Report 722108014*, RIVM, Bilthoven, The Netherlands.
- Erisman, J.W., van Pul, A. and Wyers, P. (1994) 'Parametrization of surface-resistance for the quantification of atmospheric deposition of acidifying pollutants and ozone', *Atmospheric Environment*, Vol. 28, pp.2595–2607.
- Foltescu, V.L., Pryor, S.C. and Bennet, C. (2005) 'Sea salt generation, dispersion and removal on the regional scale', *Atmospheric Environment*, Vol. 39, pp.2123–2133.
- Hanea, R.G., Velders, G.J.M. and Heemink, A. (2004) 'Data assimilation of ground-level ozone in Europe with a Kalman filter and chemistry transport model', *Journal of Geophysical Research*, Vol. 109, D10302, doi:10.1029/2003JD004283.
- Hass, H., van Loon, M., Kessler, C., Stern, R., Matthijssen, J., Sauter, F., Zlatev, Z., Langner, J., Foltescu, V. and Schaap, M. (2003) 'Aerosol modelling: results and intercomparison from European regional – scale modelling systems', *Special Report EUROTRAC-2 ISS*, Munchen.
- Ilyin, I. and Travnikov, O. (2005) 'Modelling of heavy metal airborne pollution in Europe: evaluation of the model performance', *EMEP/MS-CHE Technical Report 8/2005*.
- Jacobs, C.M.J. and van Pul, W.A.J. (1996) 'Long-range atmospheric transport of persistent organic pollutants, I: description of surface-atmosphere exchange modules and implementation in EUROS', *Report 722401013, National Institute of Public Health and Environmental Protection (RIVM)*, Bilthoven, The Netherlands.
- Kerschbaumer, A. and Reimer, E. (2003) 'Preparation of meteorological input data for the RCG-model', *UBA-Report 299 43246*, Free University Berlin Institute for Meteorology (in German).
- Liu, M.K. and Durran, D. (1977) *Development of a regional air pollution model and its application to the Northern Great Plains*, US-EPA (EPA-908/1-77-001).
- Logan, J. (1999) 'An analysis of ozonesonde data for the troposphere, recommendations for testing 3-D models and development of a gridded climatology for tropospheric ozone', *Journal of Geophysical Research*, Vol. 104, No. 16, pp.16115–16149.
- Matthijssen, J., Sauter, F.J. and de Waal, E.S. (2002) 'Modelling of particulate matter on a European scale', in J. Keller and S. Andreani-Aksojoglu (Eds). *Proceedings of GLOREAM Symposium*, 2001, Switzerland: Wengen.
- Monahan, E.C., Spiel, D.E. and Davidson, K.L. (1986) 'A model of marine aerosol generation via whitecaps and wave disruption', in E.C. Monahan and G. Mac Niocaill (Eds). *Oceanic Whitecaps and Their Role in Air/Sea Exchange*, Norwell, MA: D. Reidel, pp.167–174.

- Nenes, A., Pilinis, C. and Pandis, S.N. (1999) 'Continued development and testing of a new thermodynamic aerosol module for urban and regional air quality models', *Atmospheric Environment*, Vol. 33, pp.1553–1560.
- Poppe, D., Andersson-Sköld, Y., Baart, A., Bultjes, P.J.H., Das, M., Fiedler, F., Hov, O., Kirchner, F., Kuhn, M., Makar, P.A., Milford, J.B., Roemer, M.G.M., Ruhnke, R., Simpson, D., Stockwell, W.R., Strand, A., Vogel, B. and Vogel, H. (1996) 'Gas-phase reactions in atmospheric chemistry and transport models: a model intercomparison', *Eurotrac Report. ISS*, Garmisch-Partenkirchen.
- Quass, U., Kuhlbusch, T., Koch, M., Fissan, H., Schmidt, K-G., Bruckmann, P., Pfeffer, U., Gladtko, D. and Zang, T. (2004) 'Identification of source groups for fine dust', *IUTA Report LP 15/2004*, Duisburg, Germany: IUTA.
- Reynolds, S., Roth, P. and Seinfeld, J. (1973) 'Mathematical modelling of photochemical air pollution', *Atmospheric Environment*, Vol. 7, pp.563–598.
- Roemer, M., Beekmann, M., Bergström, R., Boersen, G., Feldmann, H., Flatøy, F., Honore, C., Langner, J., Jonson, J-E., Matthijsen, J., Memmesheimer, M., Simpson, D., Smeets, P., Solberg, S., Stern, R., Stevenson, D., Zandveld, P. and Zlatev, Z. (2003) 'Ozone trends according to ten dispersion models', *Special Report EUROTRAC-2 ISS*, Munchen.
- Schaap, M., Denier van der Gon, H.A.C., Dentener, F.J., Visschedijk, A.J.H., van Loon, M., Ten Brink, H.M., Putaud, J-P., Guillaume, B., Liouise, C. and Bultjes, P.J.H. (2004a) 'Anthropogenic black carbon and fine aerosol distribution over Europe', *Journal of Geophysical Research*, Vol. 109, D18201, doi: 10.1029/2003JD004330.
- Schaap, M., Müller, K. and ten Brink, H.M. (2002) 'Constructing the European aerosol nitrate concentration field from quality analysed data', *Atmospheric Environment*, Vol. 36, No. 8, pp.1323–1335.
- Schaap, M., van Loon, M., ten Brink, H.M., Dentener, F.D. and Bultjes, P.J.H. (2004b) 'Secondary inorganic aerosol simulations for Europe with special attention to nitrate', *Atmospheric Physical Chemistry*, Vol. 4, pp.857–874.
- Schaap, M., Roemer, M., Sauter, F., Boersen, G., Timmermans, R. and Bultjes, P.J.H. (2005) *LOTOS-EUROS Documentation, TNO Report B&O 2005/297*, TNO, Apeldoorn, The Netherlands.
- Schell, B., Ackermann, I.J., Hass, H., Binkowski, F.S. and Ebel, A. (2001) 'Modelling the formation of SOA within a comprehensive air quality modelling system', *Journal of Geophysical Research*, Vol. 106, pp.28275–28293.
- Simpson, D., Fagerli, H., Jonson, J.E., Tsyro, S., Wind, P. and Tuovinen, J-P. (2003) 'Transboundary acidification, eutrophication and ground level ozone in Europe, Part 1: unified EMEP model description', *EMEP Report 1/2003*, Norwegian Meteorological Institute, Oslo, Norway.
- Stolbovoi, V. and McCallum, I. (2002) *CD-ROM 'Land Resources of Russia'*, International Institute for Applied Systems Analysis and the Russian Academy of Science, Austria: Laxenburg.
- Travnikov, O. and Ilyin, I. (2005) 'Regional model MSCE-HM of heavy metal transboundary air pollution in Europe', *EMEP/MSCE-E Technical Report 6/2005*.
- van Loon, M. (1995) 'Numerical smog prediction II: grid refinement and its application to the Dutch smog prediction model', *CWI Research Report*, NM-R9523, ISSN 0169-0388, The Netherlands: Amsterdam, Available at: <http://www.cwi.nl/static/publications/reports/NM-1995.html>.
- van Loon, M., Bultjes, P.J.H. and Segers, A. (2000) 'Data assimilation of ozone in the atmospheric transport chemistry model LOTOS', *Environmental Modelling and Software*, Vol. 15, pp.603–609.
- van Loon, M., Roemer, M. and Bultjes, P.J.H. (2004) 'Model intercomparison in the framework of the review of the unified EMEP model', *TNO-Report R 2004/282*.

- Veldt, C. (1989) *The use of biogenic VOC-emissions*, TNO report pp.89–306.
- Visschedijk, A.J.H. and Denier van der Gon, H.A.C. (2005) 'Gridded European anthropogenic emission data for NO<sub>x</sub>, SO<sub>x</sub>, NMVOC, NH<sub>3</sub>, CO, PPM10, PPM2.5 and CH<sub>4</sub> for the year 2000', *TNO-Report B&O-A R 2005/106*.
- Walcek, C.J. (2000) 'Minor flux adjustment near mixing ratio extremes for simplified yet highly accurate monotonic calculation of tracer advection', *Journal of Geophysical Research*, Vol. 105, D7, pp.9335–9348.
- Weijers, E.P., Otjes, R.P., van Loon, M., Schaap, M., Erisman, J.W., Slanina, J., van den Bulk, W.C.M., Fonteijn, P.B., Möls, J.J. and Vermeulen, A.T. (2002) 'Concentrations, size and origin of particulate matter in the Netherlands', *ECN Report ECN-CX--02-010*, ECN, Petten, The Netherland.
- Whitten, G., Hogo, H. and Killus, J. (1980) 'The carbon bond mechanism for photochemical smog', *Environmental Science and Technology*, Vol. 14, pp.14690–14700.
- Yamartino, R.J., Flemming, J. and Stern, R.M. (2004) 'Adaption of analytic diffusivity formulations to eulerian grid model layers finite thickness', *Twenty Seventh ITM on Air Pollution Modelling and its Application*, Banff, Canada, 24–29 October.