

# The impact of emissions from the Šoštanj thermal powerplant on winter SO<sub>2</sub> pollution in Central Europe

*Danijel Čemas<sup>1</sup> and Jože Rakovec<sup>2</sup>*

<sup>1</sup> Environmental Agency of the Republic of Slovenia, Ljubljana, Slovenia

<sup>2</sup> University of Ljubljana, Faculty of Mathematics and Physics, Chair of Meteorology, Ljubljana, Slovenia

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The simulation of the wintertime transport of sulphur dioxide based on emissions made in three winter months from December 1999 to February 2000 by the thermo electric power plant located in Šoštanj (TEŠ) in Slovenia is presented. Our study is focused on estimation the regional distribution of SO<sub>2</sub> from this source – the range and level of its impact on Central Europe. The dispersion model MEDIA (Piedelievre et al., 1990), coupled with the meteorological fields of Aladin/LACE (Janoušek 1999), and the operational model for weather forecasting over Central Europe were used. The simulation was run on all winter days of the simulation period. The daily results were accumulated and averaged into monthly and seasonal estimates of air pollution caused by emissions from the TEŠ. As expected, their biggest effects are seen in the nearest regions. Despite the relatively high emission levels, high concentrations with damaging effects are mainly limited to regions approximately 50 km from the source. Slightly increased pollution levels, distinguishable from the background threshold, also spread across other parts of Slovenia, southern parts of Austria, northern parts of Croatia, western Hungary and north-eastern Italy. The TEŠ's contribution to air pollution in selected neighbouring cities is estimated. On a regional scale, the results of deposition are compared with an LADM evaluation (Berge and Jakobsen, 1998).

*Keywords:* ALADIN mesoscale meteorological model, Eulerian dispersion model MEDIA, thermal powerplant, sulphur dioxide, winter 1999/2000

## 1. Introduction

Producing around 750 MW, the Šoštanj thermo electric power plant (TEŠ) is the biggest and most important electricity provider in Slovenia, annually yielding approximately 3000 GWh of electric energy. At the same time, it is the biggest single air polluter in the region with yearly emissions of some  $50 \times 10^6$  kg of SO<sub>2</sub>. Emission levels were even much higher and exceeded  $80 \times 10^6$  kg annually before 1995, when one of five blocks was fitted

with desulphurisation devices. Emission levels are expected to radically drop further once all blocks are equipped with such devices (the second block was desulphurised in 2001). As the TEŠ is a great emitter of pollutants, there have already been several studies on its impact on the environment (*e.g.* Kotnik et al., 2000).

Despite the lower emission levels seen in Slovenia in recent years, sulphur dioxide remains one of the most important and toxic compounds in the air. A network of measurements has been established in the last few decades, and archives and annual reports showing data on emissions and air pollution are available (*e.g.* Planinšek et al., 1996, 1997 etc.). There are two monitoring systems that offer air-quality data near the ground around TEŠ. The local Environmental Information System (EIS) covering the area close to the power plant consists of six fixed automatic stations. It performs continuous monitoring of the ambient concentrations of the main air pollutants ( $\text{SO}_2$ ,  $\text{NO}_x$ ,  $\text{O}_3$  and CO) and of meteorological parameters (temperature, relative moisture and wind). Another nation-wide system ANAS (Analytical Inspection and Alarming System) monitors with a coarser spatial resolution the air quality parameters across Slovenia.

Regional modelling of air pollution dispersion helps to evaluate the extent of influence, the level of air pollution, and the amount of deposited pollutants around sources. Many of these models use the so-called Eulerian approach: a dispersion model, coupled with a meteorological model, describing the wind and dispersion properties of the atmosphere. Most of them use a limited-area meteorological model for the meteorological part, for example, the MM5 (*e.g.* Chang et al., 2000), eventually coupled with a mass-consistent model (*e.g.* Villasenor et al., 2001). Frequently, certain operational models are applied (such as Kangas and Syri, 2002; Langmann, 2000). For meteorological guidance we use the operational model Aladin/LACE (Janoušek, 1999). Some authors also use nudging of the observed wind characteristics instead of a complete meteorological model (*e.g.* Hurley et al., 1996). On larger scales or with coarser resolutions the effect of advection overcomes the importance of turbulent diffusion, being in such a case also ignored (*e.g.* Galperin and Sofiev, 2000).

Some studies are devoted to a climatological description of larger areas, thereby taking into account all emissions contributing to the pollution of an area (*e.g.* Christensen, 1997, Galperin and Sofiev, 2000). Some are devoted to specific, shorter episodes of increased pollution (Hurley et al., 1996, Langmann, 2000, Chang et al., 2000). The purpose of our model simulation is to trace the dispersion of sulphur dioxide emitted from one source, the TEŠ in Slovenia, with the aim of obtaining an estimate of its daily, monthly and/or seasonal ranges and the level of influences on a regional scale during winter-time when the air pollution problem is most problematic due to the increased electricity production and the stable weather conditions involving eventual temperature inversions of an anticyclonic weather type commonly seen in

winter across Central Europe. Accordingly, our study was carried out for three winter months (December 1999 to February 2000) to evaluate the typical winter range of pollution from the TEŠ. At the same time, it involved a test of predictions of air pollution in meso- $\alpha$  and meso- $\beta$  scales on a daily basis.

To simulate regional (medium-range) transport, diffusion and transformation or removal, an Eulerian model was applied: the French dispersion model MEDIA (Piedelievre et al., 1990). The source, advection, diffusion and sink processes were coupled with meteorological fields, forecasted operationally by the Aladin/LACE prognostic system (Aladin Int. Team, 1997, Janoušek, 1999). The output fields of simulations are three-dimensional fields of air pollution and two-dimensional wet and dry deposition fields.

## 2. Model overview

### 2.1. MEDIA

MEDIA is an Eulerian three-dimensional grid-point model in a geographical projection with a sigma vertical co-ordinate. It incorporates chemical transformations (or radioactive decays), wet and dry deposition, spatial variation of topography and weather conditions calculated from the meteorological model. Turbulence in the model is based on K-theory (Smagorinsky et al., 1965). Some modifications to the original version that were introduced are described in the following subsections.

#### 2.1.1. Dispersion equation

The concentration of the passive chemical element – the pollutant in the atmosphere – follows the mass conservation law:

$$\frac{\partial C}{\partial t} + \bar{v} \cdot \nabla C = \nabla(K\nabla C) + \dot{S}_0 + \dot{S}_i, \quad (1)$$

where  $C$  is the concentration of pollutant (strictly speaking, we use  $C$  as pollutant density:  $C = m/V$ , measured in  $\mu\text{g}/\text{m}^3$ ) at a given point in time  $t$  (s),  $\bar{v}$  (m/s) is the wind vector predicted by the numerical weather model Aladin/LACE),  $K$  ( $\text{m}^2/\text{s}$ ) is the turbulent diffusivity tensor, and  $\dot{S}_0$  and  $\dot{S}_i$  ( $\mu\text{g}/\text{m}^3\text{s}$ ) are the pollutant source and sink terms (Piedelievre et al., 1990).

To simplify the diffusion term, turbulent diffusion is modelled using only dominant diagonal terms of the turbulent diffusivity tensor:

$$\nabla(K\nabla C) = \frac{\partial}{\partial x} K_x \frac{\partial C}{\partial x} + \frac{\partial}{\partial y} K_y \frac{\partial C}{\partial y} + \frac{\partial}{\partial z} K_z \frac{\partial C}{\partial z} \quad (2)$$

The vertical diffusion coefficient is estimated with the use of the relatively simple closure approach (e.g. Louis, 1979):

$$K_z = l^2 \frac{\partial |\bar{v}_h|}{\partial z} F(Ri), \quad (3)$$

where  $\bar{v}_h$  is the horizontal wind,  $l$  is the mixing length and  $F(Ri)$  is a semi-empirical function of a Richardson number:

$$l = \frac{kz}{(1 + kz/150)}, \quad k = 0.4, \quad (4)$$

$$F(Ri) = \frac{1}{1 + 15Ri(1 + 5Ri)^{1/2}}, \quad \text{if } Ri > 0 \text{ and} \quad (5)$$

$$F(Ri) = \frac{1}{1 + 75Ri(l^2 Ri^{1/2} / (z^2 \sqrt{27}))}, \quad \text{if } Ri < 0. \quad (6)$$

where  $k$  is von Kármán constant,  $\bar{\theta}$  is potential temperature and  $g$  is gravity acceleration. The Richardson number  $Ri = \frac{g}{\bar{\theta}} \frac{\partial \bar{\theta}}{\partial z} \left( \frac{\partial |\bar{v}_h|}{\partial z} \right)^{-2}$  computation and

the form of the function  $F(Ri)$  is kept from the original French Emeraude and Peridot models (Louis et al., 1981).

In the original MEDIA formulation are the horizontal diffusion coefficients kept constant  $K_x = K_y = 5 \times 10^4 \text{ m}^2 \text{ s}^{-1}$ . Values for horizontal diffusion coefficients depend on horizontal resolution of the model. Originally it was set to  $10^5 \text{ m}^2 \text{ s}^{-1}$  in a model with horizontal resolution of one degree (Piedelievre et al., 1990); along with smaller grid also horizontal diffusion coefficients should be reduced. In our modification, not only the vertical but also the horizontal turbulent diffusion coefficients, dependent on stability as they may on a regional scale, vary considerably from one region to another and from one day to another. Therefore, a new formulation for horizontal turbulent coefficients was applied which implies the dependence on stability via the Richardson number  $Ri$ , based on the data of Pasquill (1961), cited in McCormac et al. (1971) or Hanna et al. (1980):

$$K_x = K_0 Ri_c \frac{1 + Ri}{Ri_c + Ri}, \quad Ri > 0 \text{ and}$$

$$K_x = K_0 \frac{1 - 2Ri}{1 - Ri}, \quad Ri < 0, \quad (7 \text{ a and b})$$

where  $Ri_c$  is critical Richardson number  $Ri_c = 0.22$ , and  $K_0 = 5 \times 10^4 \text{ m}^2/\text{s}$ .

### 2.1.2. Source term

After the pollutant is released with intensity  $Q(t)$  from the source, which in general is not at the grid point, it is on a subgrid scale spread around the source. The simplest approach to describe it is the isotropic Gaussian distribution, being dependant only on distance  $d$  from the source. Such approach is applied in original MEDIA:

$$\dot{S}_0(d) = \frac{\partial C}{\partial t}(d) = \frac{Q(t)}{2\pi\sigma_n^2 H} \exp\left(\frac{-d^2}{2\sigma_n^2}\right) \quad (8)$$

where  $Q(t)$  is the emission term ( $\mu\text{g/h}$ ),  $H$  is the vertical extension of the pollutant cloud (m) and  $\sigma_n^2$  is the horizontal area of the grid box including the source. But when the model results were tested and compared to measurements from the EIS and ANAS monitoring systems, the results obtained with the original formulation of source term were not very promising compared to the measurements. Calculated peak values and average concentrations of  $\text{SO}_2$  in model grid points were much lower than those measured at stations which are closer to the TEŠ than the model grid points. The modelled values were even a few times lower than measured. On the other hand, the model generally offers good results for larger distances – in mesoscale (Piedelievre, 1990). For more precise description of pollutant spread around TEŠ, model use subgrid description of pollutant expansion until it spreads to model points.

After seeking the reasons for this discrepancy, we changed i) the formulation of the description of the dispersion coefficients – as already described by equations 7a and 7b, and also ii) replaced the subgrid dispersion description (eq. 8), described here below. Our new formulation deals with the subgrid spread of pollutant before it reaches the surrounding grid points, from these it is than explicitly advected and dispersed on the model's grid scale. A simple Gaussian model and advection of pollutant cloud is used around the source until the pollutant cloud becomes big enough to bring the emitted amount of pollutant to the four nearest points.

When pollutant is emitted from the source, its mass centre  $\vec{r}$  is advected by wind  $\vec{v}$ :

$$\vec{r}_{t+\Delta t} = \vec{r}_t + \int_t^{t+\Delta t} \vec{v} dt \quad (9)$$

Wind field  $\vec{v}$  is taken from the prognostic model Aladin/LACE outputs, which are operationally available at  $\Delta\tau = 6h$  time intervals. As the time step of the dispersion model  $\Delta t = 270$  s is much shorter, the necessary forecasted fields are linearly interpolated in time.

As already mentioned, the pollutant is dissipated by the turbulent diffusion subgrid mixing into the air around the source. When the cloud becomes big enough the pollutant is included in the model. The criterion for this size, expressed by the dispersion  $\sigma$ :  $\sigma^2 \geq \Delta x^2$ ; it is calculated by a simple formula:  $\sigma^2 = 2K_x t$  where  $K_x$  is the horizontal turbulent coefficient and  $t$  is the time interval from the pollutant's release. All the emitted mass  $\Delta m$  of pollutant is distributed into the four model volume boxes  $V_i$  being representative for the model's four nearest grid points. The relative amounts of pollutant  $\Delta m_i$ , spread homogeneously over these volumes  $V_i$ , depend on their distances  $d_i$  from the centre of the pollutant cloud. Thus, the increases of concentration  $\Delta C_i$  in the respective grid points are:

$$\Delta C_i = \frac{\Delta m_i}{V_i} = \frac{\Delta m \exp(-d_i^2 / D^2)}{V_i}; \quad \sum_{i=1}^4 \exp\left(\frac{-d_i^2}{D^2}\right) = 1. \quad (10)$$

Normalisation of the sum of all four  $\Delta m_i$  is performed iteratively with  $d_i^2 / D^2$ , where weight  $D$  is, at the beginning of iteration, equal to  $\sigma$  and later approaches the appropriate value to guarantee that the exact amount of pollutant  $\Delta m$  is inserted into the model. With these changes, the wind and vertical stability would have a greater influence on pollutant dispersion. Our study tries to prove that the new version gives better results at the closest points to the TEŠ, where pollutant concentrations (both, peak values and averages) are better described.

### 2.1.3. Sink term

Three sink terms are used in the original MEDIA formulation. Wet deposition roughly describes dilution or catching using global coefficient of air-to-water transfer. Despite its simplicity is well adapted to the accuracy of precipitation predicted by model. Since model precipitation fluxes are only available at ground level, we assume that the thickness of precipitation layer is 3000 m and scavenging is uniformly active in this part of the atmosphere (Piedelievre et al., 1990). Wet deposition due to scavenging by precipitation is linearly dependant on the precipitation rate:

$$\dot{S}_w = -\frac{D_w}{h_p} = -\frac{C_m E P_r}{\rho_w h_p} = -k_w C_m \quad (11)$$

where  $D_w$  is the rate of wet deposition ( $\mu\text{g}/\text{m}^2\text{s}$ ),  $C_m$  is the mean average concentration in the precipitation layer ( $\mu\text{g}/\text{m}^3$ ),  $E = 10^4$  is the scavenging ratio,  $P_r$  is the rate of precipitation ( $\text{kg m}^{-2} \text{s}^{-1}$ ),  $\rho_w$  is the specific mass of water ( $1000 \text{ kg}/\text{m}^3$ ) and  $h_p$  is the thickness of the precipitation layer (constant – 3000 m);  $k_w$  ( $\text{s}^{-1}$ ) – the wet deposition coefficient is thus dependent on precipitation intensity. This term is applied in the lower model levels – up to the

3000 m height. The precipitation field is taken from the Aladin/LACE forecasts.

Dry deposition describes the uptake of pollutant at the Earth's surface by soil, water or vegetation. The dry deposition is taken as efficient throughout the layer closest to the Earth's surface. This process is modelled using a coefficient that is dimensionally equal to deposition velocity  $V_d$  in m/s (after Piedelievre et al., 1990):

$$\dot{S}_d = -\frac{D_d}{\Delta z} = -\frac{V_d C_g}{\Delta z} = -k_d C_g, \quad (12)$$

where  $D_d$  is the rate of dry deposition ( $\mu\text{g}/\text{m}^2\text{s}$ ),  $C_g$  is the concentration of pollutant in the air near the ground and  $\Delta z$  (m) is depth of the layer closest to the surface;  $k_d$  ( $\text{s}^{-1}$ ) – the dry deposition coefficient somehow depends on variation of  $\Delta z$  across the different topography. This term is applied in the lowest model's level only.

A lot of different reactions take place in the atmosphere. Yet it is possible to very roughly describe all chemical reactions proportional to the amount of pollutant:

$$\frac{\partial C}{\partial t} = -k_t C, \quad (13)$$

where transformation coefficient  $k_t = 10^{-6} \text{ s}^{-1}$  was chosen constant for winter conditions (Hanna et al., 1980). This term is used on all levels in the model.

The last three equations (11)–(13) together can be used to estimate the integral sink term of the whole vertical column of the model's atmosphere:

$$\dot{S}_i = -k_w C_m - k_d C_c - k_t C. \quad (14)$$

#### 2.1.4. Numerical formulation, input data, boundary conditions

In this study we are interested in regional dispersion over winter three-months period. We applied the dispersion model on the domain of approximately  $2200 \times 1700 \text{ km}^2$  covering Central Europe from  $2^\circ\text{E}$ ,  $40^\circ\text{N}$  at the SW corner in the Mediterranean Sea west of Sardinia to  $31^\circ\text{E}$ ,  $55^\circ\text{N}$  at the NE corner in southern Belarus (compare, for example, Figure 3). With a 12-km horizontal resolution we used a grid of  $182 \times 145$  points, while in the vertical we used 13 equidistant  $\sigma$ -levels from the ground to top of the model at the 300 hPa level. The time step was 720 s.

Dispersion model MEDIA needs coupling files from the meteorological model. In our case of modelling the transport in regional scale, and for whole winter time period, there is no need for coupling in very dense time and spatial intervals. Thus we couple with Aladin/LACE operational outputs in 6 h

time intervals. The 3-D coupling fields –  $u$  (zonal wind),  $v$  (meridional wind),  $w$  (vertical wind) and  $T$  (temperature) – are taken from 8 pressure levels (925, 850, 800, 700, 600, 500, 400, 300 hPa), while surface fields are 2-D:  $u10$  and  $v10$  (10 m wind),  $T2$  (2m temperature),  $p_s$  (surface pressure), pressure tendency, and stratiform and convective precipitation. Interpolation in space and in time is applied to adapt to the higher spatial and temporal resolution of the dispersion model.

As regards the input data on pollutant emissions, we put all five blocks of the TEŠ into a one-point source at 46.37°N, and 15.06°E. Daily emissions of SO<sub>2</sub>, which are archived at the Milan Vidmar Electro institute in Ljubljana were prepared for our use (Šušteršič, 2000a and b). Since the model runs with 120 time steps per day, the daily emissions were evenly distributed over all of these time steps. As the average emission over the three-month simulation was 163 tons of SO<sub>2</sub> per day, the typical time-step emission was thus between 1000 and 2000 kg per time step.

Since no physical law exists to prescribe the boundary condition in an open domain, some kind of extrapolation must be used to obtain such a boundary condition. In the model, open boundary conditions on the six sides of the integration area treated as outgoing fluxes according to Orlanski (1976) are implemented. Such a type of boundary conditions allows phenomena generated in the domain to pass through the boundaries without undergoing any significant distortion and without influencing the interior solution. This is a proper method for all open boundaries. In regional modelling it is also quite appropriate for lower boundary conditions (while for local processes in mountainous relief some limited reflection from the ground seems to offer more appropriate results *e.g.* Hanna, 1985).

## 2.2. Aladin/LACE

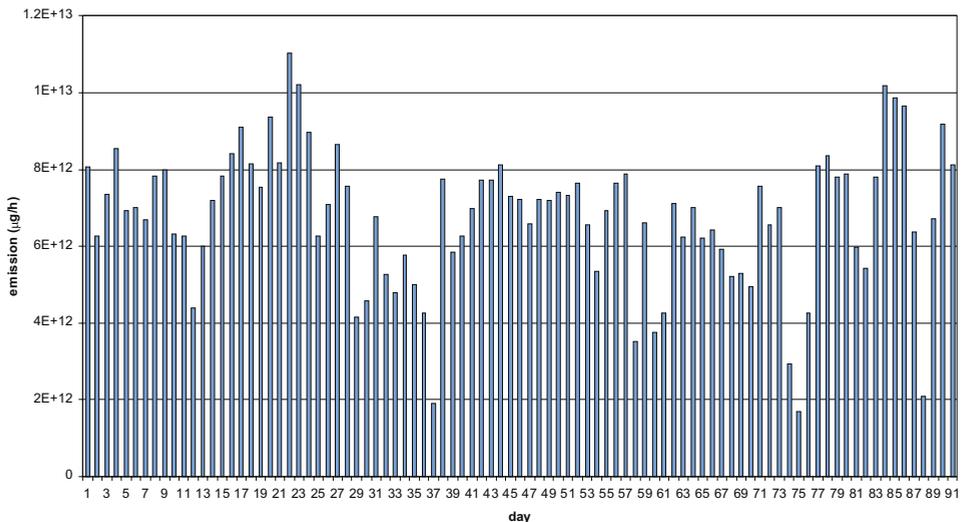
Aladin/LACE is the operational version of a limited-area weather forecasting system, one of the models being developed by the Aladin International Team (1997) from 14 countries, led by Météo-France. The operational execution of the Aladin/LACE model (Janoušek, 1999) covers an integration domain from 34.00°N, 2.18°E at the SW corner in northern Algeria, to 55.62°N, 39.0862°N at NE corner in north-western Russia. With 229 × 205 grid points, it has a horizontal resolution of 12.2 km on a Lambert map. In the vertical it has 31 hybrid levels. The time step is 568 s. It is integrated daily for a 48-hour forecasting time-range; the operational outputs are available every 3 hours. It uses initial and time-dependant boundary conditions extracted from the French current global ARPEGE forecast (every 6 h of the forecast up to 48 h) on a domain slightly larger than the Aladin/LACE integration domain (240 × 216, including the extension zone). There is no data assimilation scheme; it is run as a pure dynamic adaptation of the ARPEGE global forecast.

The main features of the model's dynamics are: hydrostatic primitive equations; the spectral method with a bi-periodic extension of the domain; a two-time level semi-Lagrangian advection scheme and semi-implicit time stepping. The main features of the physical parameterisation package are: simple radiation scheme, vertical exchange calculations taking into account a planetary boundary layer and a surface layer, shallow and deep convection, a Kessler-type large-scale precipitation scheme and an advanced surface parameterisation scheme. For a more detailed description and references, see Janoušek (1999).

### 3. The winter 1999/2000 simulation

#### 3.1. The emissions

The simulation with emissions from all five blocks together started on 1. 12. 1999 and lasted until 29. 2. 2000; a total of 91 days. The daily emissions are shown in Figure 1. The total estimated release over this period was 14,798 tons of sulphur dioxide. That means an average of 163 tons per day. The height of the highest stack is 200 m. The average exhaust volume flux of all plumes of the five blocks was 70–80 m<sup>3</sup>/s with a gas temperature of approximately 160°C enabling the plume to rise from the stacks. Plume rise depends on meteorological conditions of ambient air and plume characteristics. Typical plume rise at TEŠ is between 100 in 300 m, based on Gaussian model



**Figure 1.** Daily emissions of SO<sub>2</sub> from the TEŠ from 1.12.1999 to 29.2.2000. Emission data provided by the Milan Vidmar Electro institute in Ljubljana (Šušteršič, 2000a and b).

Screen3 (EPA, 1995). Hence, the initial cloud of pollutant was set to the effective height in the model between 200 and 350 m above the ground.

### *3.2. Regional air pollution*

Considering the resolution of the model Media, our interest was in regional scale over three-months period. We expect that over long time period and on such spatial scale conditions in PBL do not play important role in pollutant dispersion; therefore it is no need for finer time accuracy to compute diurnal effects (as, for example in Klaić, 1996). The study is not aiming in direct comparison of modeled and measured concentration close to TEŠ; it rather seeks for a larger scale »pollution climatology«.

The difference from a proper climatology lies not in the emissions, being relatively representative of winters in general, but in the particular 1999/2000 weather developments. Two different weather situations with south-west and north-east winds were essential for pollutant expansion in that winter. In December 1999 prevailing south-west winds were advecting pollutant quite a lot to the north-east; since it was also quite rainy and snowy there was a lot of a wet deposition. The opposite happened in January 2000; more north-east winds, almost no precipitation and thus more pollution in south-western regions. In February, light winds limited pollution dispersal to regions around the TEŠ and in south Slovenia. The Alps presented a high barrier for the spreading of pollutant – it almost never passed the main Alpine ridge. According to verification of Aladin/LACE precipitation field (Vivoda, 2000), the model in general overestimates precipitation over mountains (orographic precipitation) and underestimates it in lowlands. In our study case model overestimated precipitation (Table 1) which also affects wet deposition.

The results show that the pollutant SO<sub>2</sub> from the TEŠ mainly affects the regions that surround it. The average three-month elevated pollution level above the level of the natural background of 0.5 µg/m<sup>3</sup> (McCormac et al., 1971) can also be found in other regions in Slovenia, in southern Austria and north-western Croatia (Figure 2). Slightly elevated pollution levels could be also found in a belt from northern Italy towards Hungary, but only with values which could not be distinguished from the natural background. Monthly results show quantitatively quite similar results with some differences between different months (prevailing south-western flow in December, north-eastern in January, and more or less stagnant air in February).

Despite the TEŠ being large pollution source, its main environmental effect is limited to a distance of tens of kilometres around the TEŠ. As we will show in the next subsection, it also has only a low effect on air pollution in bigger Slovenian towns, contributing just 5 to 10 percent of total pollution there due to local sources (Tables 4 and 5).

Table 1. Monthly and seasonal measured and calculated precipitation (mm) and correlation coefficient  $R$  for daily precipitation comparison. (Data: Environmental Agency of the Republic of Slovenia, 2001)

Station	December 1999		January 2000		February 2000		winter 1999/2000		R
	meas.	calc.	meas.	calc.	meas.	calc.	meas.	calc.	
Ljubljana	189,0	261,1	3,9	19,8	34,6	18,2	227,5	299,1	0,51
Črna	118,7	151,9	2,4	2,4	19,4	7,7	140,5	161,9	0,80
Slovenj Gradec	104,6	139,9	4,8	7,8	25,8	6,9	135,2	154,6	0,49
Topolšica	130,6	235,1	5,6	6,4	15,3	10,4	151,5	251,9	0,77
Velenje	125,2	82,9	3,4	6,1	20,6	9,6	149,2	98,7	0,52
Žalec	133,2	228,6	4,0	14,7	31,9	21,7	169,1	265,0	0,51

Figure 3 shows the maximum concentrations of  $\text{SO}_2$  over the three-month period. Maxima were of course reached in different regions in different days of the whole winter period. Occasional, short-term stronger influences on air quality can be recognised especially north of the TEŠ. In southern Austria, in Carinthia, measurements in that winter showed at times an increased concentration of  $\text{SO}_2$  although no strong emission of  $\text{SO}_2$  occurred nearby. Our simulation indicates that these occasional increased pollution levels in the area could be due to emissions from the TEŠ.

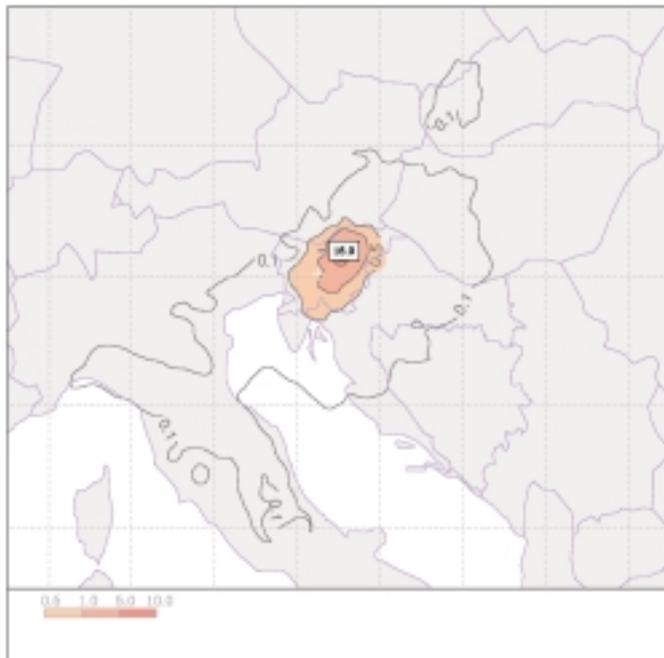
### 3.3. Wet and dry deposition

In the model the deposition of pollutant on the ground comprises wet and dry deposition. Wet deposition washes the pollutant out of the air through precipitation. The rate of wet deposition depends on the precipitation intensity and duration. During precipitation the process is relatively fast and usually washes out most pollutant in a few hours (Hanna et al., 1980). Dry deposition, on the other hand, is a uniform and continuous process, slower than wet deposition and can take several days to eliminate most of the pollutant from the atmosphere. Nevertheless, dry deposition is also an important sink for  $\text{SO}_2$  on non-precipitation days. It is estimated that on average up to 30% (Carmichael, 1984) or sometimes even up to 50% (McCormac, 1971) of  $\text{SO}_2$  is removed by this process. If precipitation intensity is low or duration of precipitation is short, wet deposition over investigated time period can be inefficient compared to the dry deposition.

The dry deposition field shows a similar pattern to the field of air pollution, while the wet deposition field is much more patterned due to the relatively irregular precipitation field. In our case, dry deposition was greater than wet deposition (Figure 4 and Table 2). Deposition had the highest intensity in December, when most precipitation occurred. As already mentioned

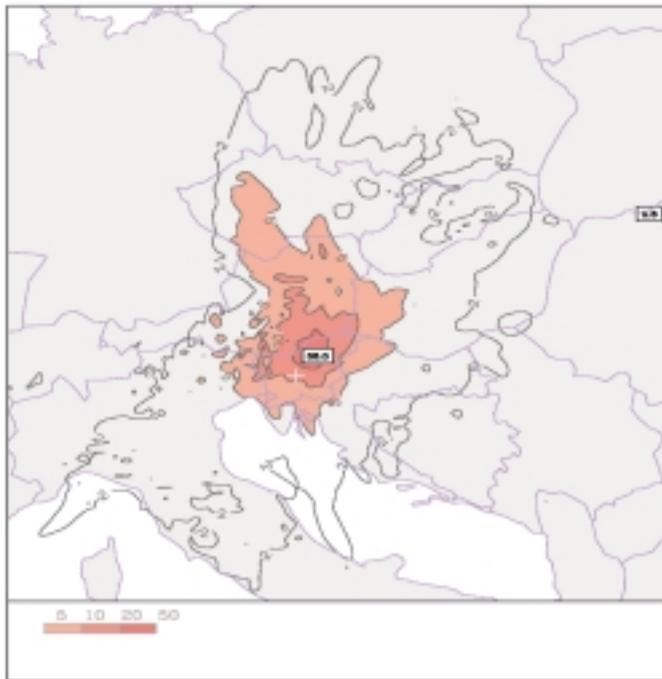
model overestimated precipitation which results in overestimation of wet deposition in most of calculated points (Table 3).

The calculated total deposition of  $\text{SO}_2$  is overestimated compared to the measured deposition of sulphur in sulphates ( $\text{SO}_4$ ), as seen in Table 3. Wet deposition depends significantly on the precipitation field which we assume was not very well forecasted by the meteorological model and is one source of error. Another source of error might be poor tuning of precipitation-scaveng-



**Figure 2.** Average modelled pollution with  $\text{SO}_2$  from the TEŠ (in  $\mu\text{g}/\text{m}^3$ ) at 850 hPa for the period from 1.12.1999 to 29.2.2000. The darker area shows the concentration of  $\text{SO}_2$  exceeding the natural background of  $0.5 \mu\text{g}/\text{m}^3$ .

ing coefficients (in equation 11). The precipitation and deposition field in the model is also more uniformly distributed around the TEŠ than in reality, due to the model's unsatisfactory resolution. However, very high total deposition was measured in Ljubljana, approximately 55 km SW of the TEŠ, compared to the level which was calculated. This is due to other sources of  $\text{SO}_2$  in Ljubljana and its surroundings.



**Figure 3.** Maximum values of pollution with  $\text{SO}_2$  at 850 hPa from the TEŠ for the period from 1.12.1999 to 29.2.2000 (not necessarily at all points simultaneously!). Gray isolines encircle those areas with more than  $2 \mu\text{g}/\text{m}^3$ , while the darker area shows where  $5 \mu\text{g}/\text{m}^3$  was exceeded at least once in the 3-month period.

#### *3.4. Influence on air quality in neighbouring cities*

We try to estimate the TEŠ's contribution to  $\text{SO}_2$  emission levels on air quality in some surrounding cities (Ljubljana, Maribor, Zagreb and Graz). Direct comparison of modeled and measured concentration from EIS-TEŠ is not suitable because of the selected resolution of the model on one hand and because of influence from local sources at these cities. Nevertheless, we look at the simulated values at the second model's level, being at approximately the effective height of the plume. The measured values, which are also strongly affected by local  $\text{SO}_2$  sources, are not directly comparable with the calculated ones and Table 3 should be considered more as a qualitative than as a quantitative comparison. As the power plant is in a basin, the second level of the neighbouring grid points is also at approximately the same altitude. However, the more distant cities are at lower altitudes. Due to the prevailing stable weather conditions, the pollutant does not spread efficiently towards the ground. Hence, pollution in the model mainly stays at this second level or higher, and maxima are generally seen at this level. It is obvious

from this table that average values according to our simulation do not contribute much, only 5 to 10%, to the air pollution in the cities that are 50 to 150 km away.

Yet it is important to mention that since pollution in the model is averaged over the volume box around the grid point, a much higher (short-term) concentration can be measured locally in some places. Thus, an inspection of individual cases (days with high pollution) shows, for example for Ljubljana, that there were two cases in December and four cases in January where pollution was over  $5 \mu\text{g}/\text{m}^3$ , which can be attributed to the TEŠ's emissions (none in February). The maximum value in the model point over Ljubljana was  $7.6 \mu\text{g}/\text{m}^3$  on 11 January 2000, when  $30 \mu\text{g}/\text{m}^3$  was measured: so one-quarter can be attributed to emissions from the TEŠ (Figure 5c).

In Maribor the  $5 \mu\text{g}/\text{m}^3$  level was exceeded once (with a value  $5.2 \mu\text{g}/\text{m}^3$  on 7 December 1999 – in comparison with the  $25 \mu\text{g}/\text{m}^3$  measured on that day). In Zagreb, only five times did the computed influences exceed  $1 \mu\text{g}/\text{m}^3$

Table 2. Cumulative wet and dry deposition of  $\text{SO}_2$  (in  $\text{mg}/\text{m}^2$ ) from 1. 12. 1999 to 29. 2. 2000, calculated in selected model points close to the source

Distance from source	dry deposition	wet deposition	total deposition
First point to NW (approx. 8 m)	220	144	364
First point to NE (approx. 6 km)	271	69	340
First point to SW (approx. 9 km)	234	154	388
First point to SE (approx. 11 km)	282	47	329
Ljubljana (approx. 55 km SW)	35	46	81
Maribor (approx. 50 km NE)	43	17	60

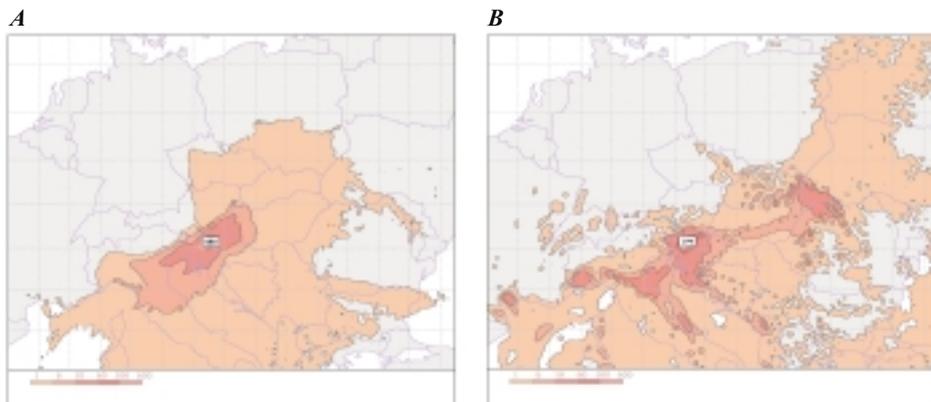


Figure 4. Calculated dry (a) and wet (b) deposition of  $\text{SO}_2$  (in  $\text{mg}/\text{m}^2$ ) from 1.12.1999 to 29.2.2000.

Table 3. Accumulated total deposition of sulphur S in sulphates (in mg/m<sup>2</sup>) measured and calculated (Data: A. Šegula, Environmental Agency of the Republic of Slovenia, 2001)

City or point	December 1999		January 2000		February 2000		Winter 1999/2000	
	meas.	calc.	meas.	calc.	meas.	calc.	meas.	calc.
first point to NW (approx. 8 m)	65	104	11	45	32	33	108	182
first point to NE (approx. 6 km)	87	71	–	56	40	43	–	170
first point to SW (approx. 9 km)	–	100	–	56	46	41	–	197
first point to SE (approx. 11 km)	66	52	–	58	36	54	–	164
Ljubljana (approx. 55 km SW)	92	28	17	8	36	3	146	40
Celje (approx. 40 km SE)	55	58	10	32	15	34	79	124

Table 4. Monthly average pollution with SO<sub>2</sub> (in µg/m<sup>3</sup>), measured in some cities and calculated for four neighbouring points on the 2<sup>nd</sup> model level. Note that TEŠ contribute only 5–10% to total pollution.

City	December 1999		January 2000		February 2000	
	meas.	calc.	meas.	calc.	meas.	calc.
Ljubljana <sup>1</sup>	18	1.1	30	1.7	19	0.9
Maribor <sup>2</sup>	38	1.5	38	1.1	24	1.2
Zagreb <sup>3</sup>	37	0.2	43	0.4	33	0.4
Graz <sup>4</sup>	15.8	0.5	16.8	0.7	11	0.3

Table 5. Maximum daily concentration of SO<sub>2</sub> (in µg/m<sup>3</sup>), measured in some cities and calculated for four neighbouring points on the 2<sup>nd</sup> model level. Note that TEŠ contribute only 5–10% to total pollution.

City	December 1999		January 2000		February 2000	
	meas.	calc.	meas.	calc.	meas.	calc.
Ljubljana <sup>1</sup>	47	5.9	67	7.6	30	4.9
Maribor <sup>2</sup>	82	5.2	75	4.9	47	4.6
Zagreb <sup>3</sup>	73	1.4	85	2.6	54	1.8
Graz <sup>4</sup>	42	7.2	29	7.0	20	6.4

Data sources for Tables 4 and 5:

<sup>1</sup> and <sup>2</sup> – Planinšek et al. 1999/2000

<sup>3</sup> – average of data for 5 different stations in Zagreb (centre, north, east, south and west) kindly provided by the Institute for Medical Research and Occupational Health of Croatia (Vadjic 2001, personal communication)

<sup>4</sup> – average of data for 5 different stations in Graz (centre, north, east, south and west), obtained from the Internet site: <http://www.hasnerpl.asn-graz.ac.at/luft/index.htm>

(with a maximum of  $2.6 \mu\text{g}/\text{m}^3$  on 26 January 2000); for comparison, the maximum measured daily value reached  $140 \mu\text{g}/\text{m}^3$  one day in January at the station »west« (Figure 5b). Graz has a relatively low average computed influence from the TEŠ, but some higher simulated peaks: seven times over  $2 \mu\text{g}/\text{m}^3$  and three times over  $5 \mu\text{g}/\text{m}^3$  (with a simulated maximum of  $7.2 \mu\text{g}/\text{m}^3$  on 24 December 1999, Fig. 5c).

The comparison of the simulated air pollution, being representative for the average pollution in the models' volume boxes, with the measured values in the points reveals several disadvantages, such as: i) topography in the model is resolved poorly and along with it the local wind conditions; ii) vertical resolution hardly resolves the eventual stable layers and temperature inversions; and, iii) the measured pollution is strongly affected by local sources.

### 3.5. Deposition over Europe

We have mentioned some of the troubles associated with validation of the model with point-measured values. Thus, we try to perform a qualitative comparison of our results across Europe with the ones of the European Monitoring and Evaluation Programme (Jacobsen et al., 1997; EMEP, 1998). The EMEP results are obtained by LADM – an Eulerian model, with  $50 \times 50 \text{ km}^2$  resolution, for calculating sources, dispersion and sinks of pollutants over Europe. In the model, a linear parameterisation scheme for dispersion, chemical processes, wet and dry depositions are used (Berge and Jakobsen, 1998, Klaić, 2003).

Direct comparison is impossible due to various differences (such as the different geometry of the two models, different resolutions, different specifications of sources etc.). Accordingly, for a qualitative estimation only, a recalculation of our results was needed for the LADM's characteristics. Also, as the LADM deals with annual emissions from the whole of Slovenia, our 3-month results for the TEŠ's emissions were in this sense also re-normalised. Our test study calculates diffusion of  $\text{SO}_2$  from one point source – TEŠ – compared to LADM which considers all  $\text{SO}_2$  sources in Slovenia. Since TEŠ contributes 47% of total  $\text{SO}_2$  emission in Slovenia in 1996 (Planinšek et al., 1997) and for that representative source of  $\text{SO}_2$  in Slovenia, the comparison between the two calculations seems reasonable. Nevertheless, some aspects cannot be recalculated or re-normalised to become comparable. One of these is the 4-times finer horizontal spatial resolution of MEDIA, resulting in much higher Alps than those represented in the LADM. That makes this mountain ridge a bigger obstacle to pollutant dispersion in MEDIA.

The main difference between the results of the two models is caused by the specific precipitation field of the 1999/2000 winter. In the LADM a statistical representation (ten-year average) for precipitation was used, while in MEDIA a forecasted precipitation field (for 24 h in advance) was used for the specific winter of 1999/2000.

During some anticyclonic situations over Central Europe involving easterly winds across Slovenia, the deposition field spread more to the south-west, while in the averaged meteorological conditions over ten years (LADM) such advection is not very frequent; thus the results of LADM do not show the impact of emissions from Slovenia towards the south-west.

A comparison shows that the maximum of MEDIA is greater than the maximum of the LADM, which can be attributed to the better resolution and point source in MEDIA. Taking all of the mentioned restrictions on the comparison into account, the results and correspondence of the two models are still reasonable.

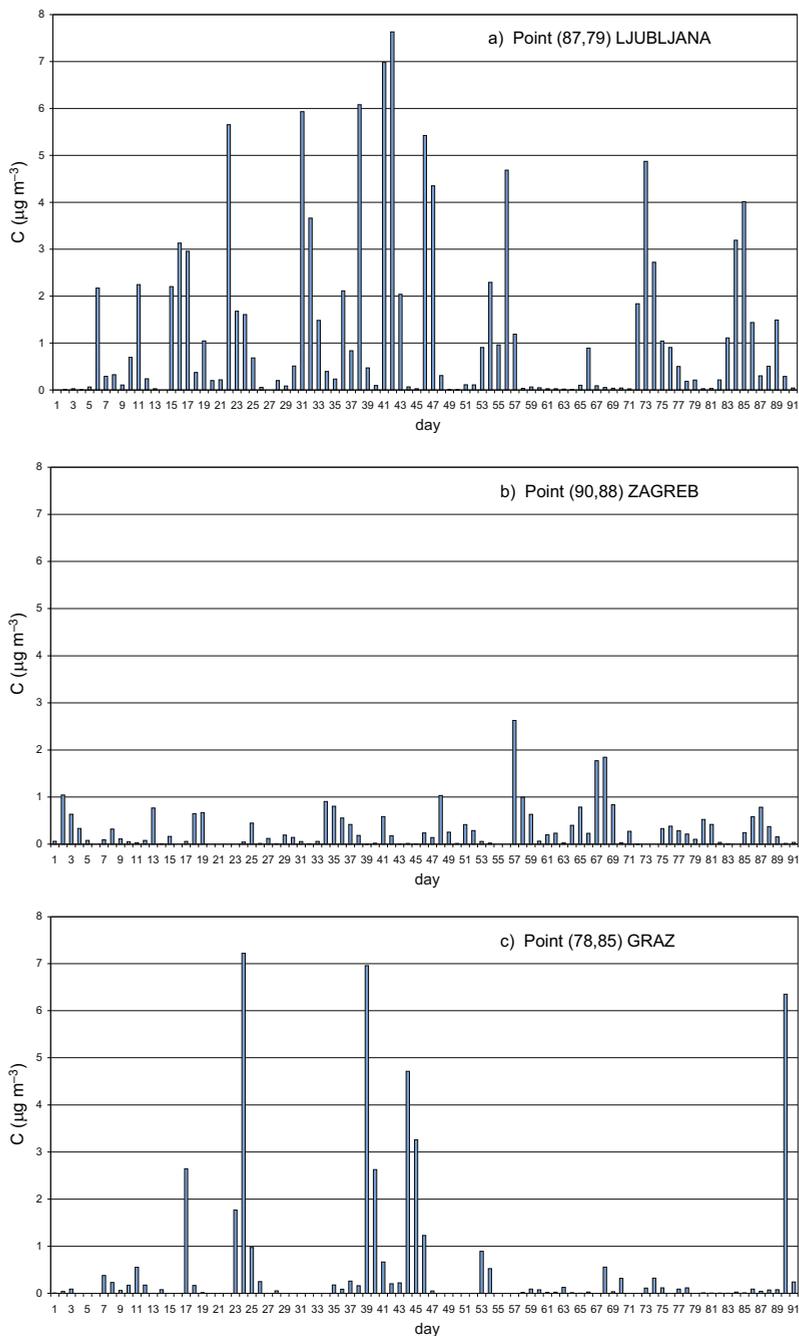
#### 4. Conclusions

An estimate of air pollution in Central Europe in the winter of 1999/2000 caused by emissions of SO<sub>2</sub> from the TEŠ was obtained by a simulation using the numerical model MEDIA, coupled with operationally forecasted meteorological fields. Two different weather situations with south-west and north-east winds were essential for pollutant advection and dispersion in that winter. The Alps generally represent a significant barrier to the spread of pollutant towards the north.

Despite the relatively strong emissions from the TEŠ, the main pollution effects are limited to a roughly ten-kilometre range around the TEŠ. Regarding the average for the whole winter, a smaller amount of sulphur dioxide, at the level of the natural background, can also be found in other regions of Slovenia, in southern parts of Austria and in north-western Croatia. Short-term impacts on air quality can be expected mainly north of the TEŠ. Measurements in remote places in southern Austria, excluding local emissions, support our model results that any eventual increase in pollution in this area could be due to emissions from the TEŠ.

Any direct evaluation of the model results, being representative for model-grid volumes with point-measured ones, is difficult for several restricting factors. The relatively complex mountainous topography in the model is resolved only partly; thus local winds are also not very precise. Vertical resolution hardly resolves the eventual atmospheric stable layers and temperature inversions. On the other hand, the measured pollution values are strongly affected by local sources. Considering all of these restrictions, the effect of the TEŠ on air pollution in bigger cities in the area, averaged over three winter months, is estimated to be just 5 to 10 percent of total pollution measured there.

More reliable is the comparison of area-averaged values of depositions with those ones of another model: the LADM. Although this one uses climatological information and in our simulation the daily forecasts of one particular winter, the agreement shows some common features.



**Figure 5.** A 24-hour concentration of  $\text{SO}_2$  on  $2^{\text{nd}}$  model level due to emissions from the TEŠ in the model point over Ljubljana, Zagreb and Graz from 1.12.1999 to 29.2.2000.

As better local forecasts of air pollution could be expected when using the better resolution of both the meteorological and dispersion models, thus a more accurate topography representation and its influence on meteorological fields could also be anticipated. What is currently not yet well forecasted is the channelling of the winds in the valleys, inversion layers and other local meteorological phenomena having a direct impact on air pollution and acid rain. One possible solution in this direction is to use meteorological data from the dynamic adaptation of the Aladin/SI model with a 2.5-km resolution (Žagar and Rakovec 1999, Žagar 2000). Improved model resolution would also make verification with point-measured values more reliable. While achieving some progress in this direction, including the chemical part of the model, in the version used the relatively rudimentary resolution should be improved.

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## References

- Aladin International Team (1997): The ALADIN project: mesoscale modelling seen as a basic tool for weather forecasting and atmospheric research. *WMO Bull.*, **46**, 317–324
- Berge, E. and H. Jakobsen, A. (1998): A regional scale multi-layer model for the calculation of long-term transport and deposition of air pollution in Europe. *Tellus*, **50**, 205–223.
- Carmichael, G. R. and Peters, L. K. (1984): An Eulerian Transport/Transformation/Removal Model for SO<sub>2</sub> and Sulphate – I. Model Development. *Atmospheric Environment*, **18**, 937–951.
- Chang, K. H., Jeng, F.-T., Tsai, Y.L. and Lin, P.L. (2000): Modelling of long range transport on Taiwan's acid deposition under different weather conditions. *Atmospheric Environment*, **34**, 3281–3295.
- Christensen, J. H. (1997): The Danish Eulerian Hemispheric model – a three-dimensional air pollution model used for the Arctic. *Atmospheric Environment*, **31**, 4169–4197.
- EMEP (1998): Transboundary Acidifying Air Pollution in Europe. EMEP **7/98**, MSC-W; Oslo, NMI.
- EPA (1995): SCREEN3 Model's Users Guide, EPA-454/B-95-004, Office of Air Quality Planning and Standards, Research Triangle Park, N.C., September 1995.
- Galperin, M. V. and Sofiev, M. A (1998): The long-range transport of ammonia and ammonium in the northern hemisphere. *Atmospheric Environment* **32**, 373–380.

- Gjerek, M. (1999, 2000): Air Pollution, Mesečni bilten (Monthly bulletin) HMZ **6/12**, Ljubljana, 50–6, HMZ **7/1**, Ljubljana, 41–53, HMZ **7/2**, Ljubljana, 48–59.
- Graz SO<sub>2</sub> data: <http://www.hasnerpl.asn-graz.ac.at/luft/index.htm>.
- Hanna, S. R., Briggs, G. A. and Hosker, R. P., Jr., (1980): Handbook on Atmospheric Diffusion. Oak Ridge, Tennessee, Technical Information Center – U. S. Department of Energy. pp. 25–35, 67–73.
- Hanna, S. R. (1985): Air quality modelling over short distances. In: D. D. Houghton (Ed.): Handbook of applied meteorology. New York etc. Wiley Interscience Publ., pp. 712–743.
- Hurley, P. J., Manins, P. C. and Noonan, J. A. (1996): Modelling wind fields in MAQS. Environmental Software **11**, 35–44.
- Jakobsen, H. A., Jonson, J. E. and Berge, E. (1997): The multi-layer Eulerian model: Model description and evaluation of transboundary fluxes of sulphur and nitrogen for one year. EMEP **7/97**, MSC-W; Oslo, NMI.
- Janoušek, M. (1999): ALADIN/LACE characteristics. Regional Centre for Limited Area Modelling in Central Europe, <http://www.chmi.cz/meteo/ov/lace/docs/aladin-fs.html>.
- Kangas, L. and Syri, S. (2002): Regional nitrogen deposition model for the integrated assessment of acidification and eutrophication. Atmospheric Environment (in press)
- Klaić, Z. (1996): A Lagrangian Model of Long-Range Transport of Sulphur with the Diurnal Variations of Some Model Parameters, J. Applied Meteorol. **35**, 574–586.
- Klaić, Z. B. (2003): Assessment of wintertime atmospheric input of European sulfur to the eastern Adriatic, Il Nuovo Cimento **C 26**, 1–6.
- Kotnik, J., Horvat, M., Mandić, V. and Logar, M. (2000): Influence of the Šoštanj coal-fired thermal power plant on mercury and methyl mercury concentrations in Lake Velenje, Slovenia. The Sci. of Total Env. **259**, 85–95.
- Langmann, B. (2000): Numerical modelling of regional scale transport and photochemistry directly with meteorological processes. Atmospheric Environment **34**, 3585–3598.
- Louis, J. F. (1979): A parametric model of vertical eddy fluxes in the atmosphere. Bound. Layer Meteorol., **17**, 187–202.
- Louis, J. F., Tiedtke, M. and Geleyn, J. F. (1981): A short history of the operational PBL parameterisation at ECMWF. Proc. ECMWF Workshop on PBL Parameterization, Reading, 59–79.
- McCormac, M. (ed.) and Varney, R. (1971): Introduction to the Scientific Study of Atmospheric Pollution. D. Riedel Publishing Company, Dordrecht, Holland.
- Orlanski, I. (1976): A simple boundary condition for unbounded hyperbolic flows. J. Comput. Phys., **21**, 251–269.
- Pasquill, F. (1961): The Estimation of the Dispersion of Windborne Material. Meteorol. Mag., **90**, 33–49.
- Piedelievre, J. P., Musson-Genon, L. and Bompay, F. (1990): MEDIA-An Eulerian Model of Atmospheric Dispersion: First Validation on the Chernobyl Release. J. Appl. Meteorol., **29**, 1205–1220.
- Planinšek, A. (1996, 1997 etc.): Onesnaženost zraka v Sloveniji v letu 1995(1996, etc.) (Air pollution in Slovenia in 1995 (1996 etc.)). Environmental Agency of the Republic of Slovenia, Ljubljana.
- Smagorinsky, J., Manabe, S. and Holloway, J. L., Jr. (1965): Numerical Results from a 9-Level General Circulation Model of the Atmosphere. Mon. Wea. Rev., **93**, 727–768.
- Šegula, A. (2001): personal communication.
- Šušteršič, A. (2000a): Ocena emisij snovi v zrak in rezultati meritev emisijskih koncentracij TE Šoštanj v letu 1999 (Estimate of emissions of pollutants into air and results of measured emission concentrations for TE Šoštanj in 1999). EKO 241/00, Ljubljana, EIMV.
- Šušteršič, A. (2000b): Data on emission and emissions due to TEŠ – Podatki o emisijah in imisijah iz TEŠ from 1.12.1999 to 29. 2. 2000. Ljubljana, EIMV.

- Villasenor, R., Claiborn, C., Lamn, B. and O'Neill, S. (2001): Mesoscale revision of wintertime matter episodes in eastern Washington, USA. *Atmospheric Environment* **35**, 6479–6491.
- Vivoda, J. and Bartok, J. (2000): Verification of precipitation predicted by Aladin/LACE and Aladin/Slovakia. *Aladin Newsletter* 19. July – December 2000.
- Vadjjić (2001): Zagreb SO<sub>2</sub> data: Institute for Medical Research and Occupational Health of Croatia, personal communication.
- Žagar, M. and Rakovec, J. (1999): Small-scale surface wind prediction using dynamic adaptation. *Tellus* **51A**, 489–504.
- Žagar, M. (2000): Napovedovanje pojavov majhnih razsežnosti z dinamičnim prilagajanjem: disertacija v somentorstvu – Adaptation dynamique pour les prévisions de phénomènes météorologique a echelle tres fine: these en co-tutelle. Ljubljana – Toulouse, 178 pp.

## SAŽETAK

## Utjecaj emisija termoelektrane Šoštanj na onečišćenje Srednje Europe sumpornim dioksidom tijekom zime

*Danijel Čemas i Jože Rakovec*

Prikazana je simulacija transporta sumpornog dioksida, koji je tijekom tri zim-ska mjeseca od prosinca 1999. do veljače 2000. emitiran iz termoelektrane Šoštanj (TEŠ) u Sloveniji. Naša studija je fokusirana na procjenu regionalne razdiobe SO<sub>2</sub>, koji potječe iz TEŠ, odnosno na razinu i raspon utjecaja TEŠ na Srednju Europu. Primijenjeni su model disperzije MEDIA (Piedelievre i sur., 1990), koji je združen meteorološkim poljima Aladin/LACE modela (Janoušek 1999), te operativni prognostički model za Srednju Europu. Simulirani su svi dani u promatranom zimskom razdoblju. Na temelju dnevnih modeliranih vrijednosti procijenjene su srednje mjesečne i sezonske vrijednosti onečišćenja uzrokovanog emisijama TEŠ. Kao što se i očekivalo, utjecaj termoelektrane je najveći u područjima najbližim izvoru. Unatoč relativno velikim razinama emisije, visoke koncentracije sa štetnim efektima uglavnom se nalaze na udaljenostima od izvora do 50 km. Blago povišene razine onečišćenja, koje je moguće razlikovati od temeljnog onečišćenja, također se nalaze u drugim dijelovima Slovenije, dijelovima južne Austrije, sjeverne Hrvatske, zapadne Mađarske i sjeveroistočne Italije. Procijenjen je i doprinos TEŠ-a onečišćenju zraka u odabranim susjednim gradovima. Procijenjena taloženja uspoređena su na regionalnoj skali s procjenama LADM modela (Berge i Jakobsen, 1998).

*KLjučne riječi:* mezoskalni meteorološki model ALADIN, Eulerov model disperzije MEDIA, termoelektrana, sumporni dioksid, zima 1999/2000

Corresponding author's address: D. Čemas, Environmental Agency of the Republic of Slovenia, Vojkova 1b, SI-1000 Ljubljana, Slovenia, e-mail: danijel.cemas@rzs-hm.si