Environment Protection Engineering

Vol. 30 2004 No. 3

KATARZYNA JUDA-REZLER*

MODELLING OF THE AIR POLLUTION WITH SULPHUR SPECIES IN POLAND

In Poland, emission of sulphur dioxide into the atmosphere is still substantial, resulting in a direct and an indirect threat to human health and the environment. This paper compares current (1999) and past (1992) SO_x concentrations and sulphur depositions calculated for Poland using a regional-scale numerical dispersion model. Annual mean concentrations of SO_2 and SO_4^{2-} as well as total sulphur deposition are calculated for selected years. The results show that air pollution with sulphur species in Poland is decreasing significantly, but at some locations the concentrations and deposition of sulphur are still high. Model results for 1999 have been verified by the data from the National Network for Air Quality Monitoring. In order to evaluate the model performance, a comprehensive package of methods was applied. The results of model performance evaluation are presented and discussed. The SO_2 concentrations measured and calculated proved to be in a very good agreement. Available measurements show that the model underestimates SO_4^{2-} concentrations but satisfactorily predicts a total sulphur deposition.

1. INTRODUCTION

Poland as the candidate for EU speeded up its development over the last decade; the changes started in the early 1990s. These include basic restructuring and modernizing industry (heavy industry reduction), the implementation of abatement technologies (desulphurisation of flue gases), introduction of a broad energy-saving programme as well as alteration of the composition of energy supplies by reducing coal consumption. Simultaneously, environmental legislation transposing EU directives, in particular relating to air quality, were adopted. All these measures led to a decrease of atmospheric emissions and to a significant improvement of the air quality. However, at some locations in Poland, air quality problems still persist.

The emissions of SO_2 into the atmosphere are directly connected with the composition of energy supplies. During the period of interest (1992–1999) coal consumption in Poland was reduced by more than 10%, from over 75% in 1992 to approximately

* Institute of Environmental Engineering Systems, Warsaw University of Technology, Nowowiejska 20, 00-653 Warsaw, Poland. E-mail: Katarzyna.Juda-Rezler@is.pw.edu.pl

65% in 1999. The amount of SO_2 emitted decreased by almost 40%, dropping from 2820 Gg in 1992 to 1719 Gg in 1999. Despite that the emission in 1999 represented almost 10% of the European emissions. These numbers show that substantial reductions in SO_2 emissions have been achieved, but they are still significant. In Poland, main sources of air pollution are the following: power generation, industry production, municipal sector and transport, and power generation whose contribution to the total SO_2 emission approaches 50% (see EMEP [8]). Air pollution with sulphur species is considered, since – in contrary to the majority of European countries – SO_2 emissions into the atmosphere and its impact on human health as well as on natural and man-made environments in Poland are still considerable.

For these reasons the tools for assessing the air pollution as well as the resulting risk to human health and the environment are needed. To this end intensive studies have been carried out at the Warsaw University of Technology. For sulphur species an integrated assessment model ROSE (risk of airborne sulphur species to the environment) has been developed for the area of Poland. The sensitivity of environment to the sulphur species was assessed by calculating and mapping of critical thresholds (above which damage may occur), i.e. the critical levels for SO_2 and SO_4^{2-} and the so-called *conditional critical loads* being responsible for an acidifying effect of sulphur. The ratios of current levels/loads to critical ones were calculated and the areas at risk were specified (JUDA-REZLER [13]). As a part of the ROSE model, the POLSOX-II numerical air-pollution model was applied in the dispersion calculations. First results, mean annual SO_2 concentrations, have already been presented (JUDA-REZLER [14]).

Air-pollution modelling in Poland has been carried out for the last 20 years, including, among others, the urban scale SO_2 modelling (JUDA [12]), the regional scale SO_x and NO_x modelling (ABERT et al. [1]), modelling for the regulatory purposes (MARKIEWICZ [21]), the regional and long-range transport of heavy metals modelling (BARTNICKI [4], BARTNICKI et al. [5], ULIASZ et al. [22]) and, recently, multi-scale oxidants modelling (KAMIŃSKI et al. [17]).

This paper is focused on air pollution modelling for the task of its integrated assessment. The aim was to demonstrate the current (1999 data) state of the air pollution in the country compared to the past (1992 data) state. The POLSOX-II model's comprehensive results for two selected years as well as model performance evaluation are presented and discussed.

2. MODELLING

2.1. THE STUDY AREA

The study area for the national scale modelling is configured to be bigger than the area of a given country, because the inflow of pollutants from the outside of the country's boundaries needs to be taken into account. Therefore, the accepted study area of 1050 km

× 900 km includes Poland and parts of the neighbouring central European countries. Two computational grids were used, both being a part of the "Cooperative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe" (EMEP) model grid, but with finer spatial resolution (30 km × 30 km). The first, the so-called outer grid, covers the entire study area. The second smaller grid (900 km × 750 km) covers the area of Poland and is used only for the presentation of the modelling results (see figure 1).

2.2. MODEL DESCRIPTION

The POLSOX-II is the newest type of numerical two-dimensional Eulerian grid model of the well-mixed layer dedicated to sulphur species, developed at the Warsaw University of Technology (ABERT et al. [1], JUDA-REZLER and ABERT [15], LOWLES et al. [20]). The model code combines transport, diffusion, chemistry and deposition processes (dry and wet) in the study area. When developing the model the following assumptions were made: the model should be operational, relatively simple, supported by readily available input data, computationally fast and adequate for long-term regional scale calculations. In order to meet these requirements, the model code contains necessary simplifications.

In the model, advection–diffusion (K theory) equations (with removal term including dry and wet deposition as well as chemical transformation) are numerically solved by means of the two-cyclic Galerkin finite-element method with linear basis functions (ZIENKIEWICZ and TAYLOR [25]) for SO_2 and SO_4^{2-}. The time derivatives are approximated by the Crank–Nicholson method, and a method of Fractional Steps (YANENKO [24]) is used to expand the algorithm to two dimensions. The negative values in the solution are eliminated by the global filter (BARTNICKI [3]). The advection–diffusion portion and the removal portion of the basic equations are separated. The advection–diffusion portion is solved with the time step equal to 0.5 h, while the removal portion is solved with the time step equal to 120 s. As the accepted so-called outer computational grid is much wider than Polish territory, boundary conditions are set at zero.

Meteorological fields are prepared by meteorological pre-processor EASEMET (JUDA-REZLER and ABERT [15]) from ECMWF (European Centre for Medium-Range Weather Forecasts Reading, UK) source data with 0.5 deg × 0.5 deg resolution (data sets four times a day). Advection wind is obtained as an average wind in the mixing layer – extrapolated based on the surface wind (interpolated for each data set based on the source data) and objectively analysed. Precipitation, temperature and cloudiness fields are interpolated (by the method of weight interpolation) based on the source data (for each data set, precipitation for each 12 hours). Stability-dependent, variable mixing layer height is calculated for each data set.

In the removal, submodel constant dimensionless scavenging ratios are used. The transformation of SO_2 to SO_4^{2-} is parameterised as the first-order reaction with transformation rate having sinusoidal variation over the year with the maximum value at

the summer solstice. A simple submodel of deposition is implemented. It allows us to calculate the velocities of SO_2 dry deposition and the parameters of local dry deposition in the study area. The velocity of dry deposition of SO_4^{2-} is kept constant. The parameter of local dry deposition determines the proportion of the total amount of pollutant emitted within a single grid cell and deposited in the same grid cell (this pollutant does not undergo transport and diffusion processes). The parameter is a function of the atmosphere stability, wind velocity, height of emission source, dry deposition velocity and computational grid step and ranges between 0.0 and 0.05. Space- and time-dependent velocities (V_d) of SO_2 dry deposition are calculated for the constant height of flux layer (being approximately equal to 50 m), using resistance analogy formula and the Monin–Obukhov similarity theory for the surface layer. The basic $V_d(SO_2)$ referring to the height $z = 1$ m was 0.7 cm s^{-1}. The Businger–Dyer integrated stability functions are applied to the calculations of aerodynamic resistances. The Monin–Obukhov length for each time-step (6 h) was determined using GOLDER'S relationships [9], which take an average roughness length for the study area and the Pasquill stability class, established based on meteorological data.

The model operates at a time step of six hours (time interval for time-dependent data input) and is used for calculations of seasonal or annual values. However, the time scale of the results obtained can be smaller and can constitute any multiple of the basic time step. As a result, the concentration matrices for SO_2 and SO_4^{2-} as well as total (dry and wet) sulphur deposition in the area covered with the computational grid are obtained.

2.3. MODEL INPUT DATA

In order to compare present and past air quality in Poland, model input data were collected for two base years:

- 1992 – representing past pollution in Poland,
- 1999 – representing current pollution in Poland.

Emission data from Poland as well as from parts of east Germany and the Czech Republic were collected for two selected years. An inventory of SO_2 emission for the Czech Republic and for eastern Germany came from EASE project database (LOWLES et al. [20]). This database contains an official inventory of emission for the Czech Republic (REZZO database) and for eastern Germany for 1992 as well as the official "prognosis for the year 2000" for these countries. The prognostic data were considered to represent 1999 emissions. An emission inventory for Poland came from a database existing at Warsaw University of Technology. There are several categories of sources in this database which includes: public power generation, industrial energy generation and industrial production processes and the municipal sector. Data were gathered for 1992 and for 1999. All together the emission database consists of 2953 (in 1992) and 2908 (in 1999) point sources and 1627 area sources. For the coordinates of point sources yearly emission and source parameters are given. For area sources grided annual totals are

prepared. A seasonal adjustment is then applied to public power generation and municipal sources, setting an expected monthly proportion of the total.

As the modelling area covers parts of neighbouring countries, national meteorological data are insufficient. In the EASE project, the observational and analysed 1992 meteorological data (for hours 00, 06, 12 and 18) from ECMWF (Reading, UK) were used. The data contained wind speed and direction, temperature, precipitation and cloud cover, surface roughness, u- and v-stress. Meteorological fields necessary for the POLSOX-II model were created by the meteorological pre-processor EASEMET. The Pasquill stability classes (based on surface wind speed, daytime insolation and cloud cover) were also determined for each of the ECMWF original grid data representing each data set. Based on analysed observations of temperature profiles throughout Poland in the period of 1966–1980 (from four radiosouding stations), mean values of the mixing height (H) were estimated for each Pasquill stability class (LITYŃSKA [18]). The resulting heights of mixing layer for the Pasquill stability classes calculated in each time step of computation are used.

Unfortunately, ECMWF data for 1999 was not available. However, a comprehensive analysis showed that for long-term calculations the standard meteorological data could be applied, which introduced relatively small error. First of all the meteorological conditions within the modelling area for both years were compared (based on hydrometeorological bulletins). West circulation was responsible for creation of meteorological conditions; anticyclone stagnations approached the multi-year average, temperature exceeded the multi-year average and total precipitation was below the multi-year average. Secondly, the influence of interannular variability of meteorological input fields on the concentration and deposition values predicted based on the Eulerian grid model (using the same emission data) was previously investigated (ABERT et al. [2]). The meteorological fields prepared based on source data for five successive years (1988–1992) were examined. It turned out that annually averaged modelling results varied from year to year to a little degree when the same model code and emission data are employed. The influence of meteorological fields was found to be small and insignificant, both in respect of concentration/deposition distribution and values. The differences between both annual average SO_2 concentrations and total sulphur deposition did not exceed 6% and 5.2%, respectively. Taking the above considerations into account, the calculations for both years were made using 1992 meteorological data.

3. RESULTS AND DISCUSSION

The results of the POLSOX-II model run for 1992 and 1999 are presented as the concentration of sulphur species and deposition maps. Annual average concentrations of sulphur dioxide and sulphate in the air are shown in figures 1 and 2, respectively. Annual depositions of sulphur are given in figure 3. Predicted annual average concentrations of sulphur species in the air and deposition rates of total sulphur averaged

over the Polish territory for years 1992 and 1999 are shown in table 1. They indicate that, in general, concentration and deposition are reduced by approximately a factor of 2 over the period.

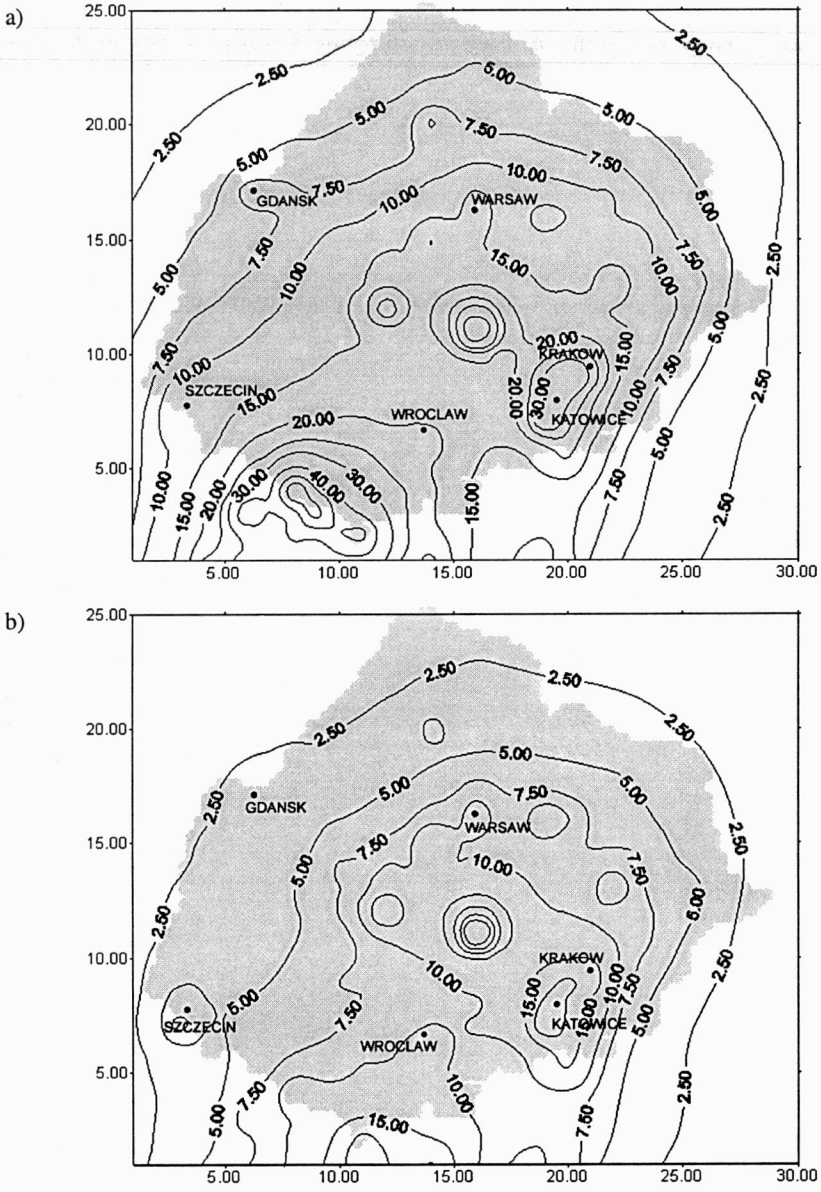

Fig. 1. Calculated yearly averaged SO_2 concentrations in Poland [μg (SO_2) m^{-3}] in 1992 (a) and in 1999 (b), JUDA-REZLER [16]

On the maps of calculated annual mean SO_2 concentration (figure 1), one can clearly see the distribution of emission sources in Poland and surroundings countries, especially Germany and the Czech Republic. In 1992, three large areas with high sulphur concentrations can be separated. The first one is located in the so-called 'Black Triangle' Region (boundary of Poland, the Czech Republic and Germany), where the largest sources of anthropogenic emissions of sulphur dioxide are found, e.g. lignite power stations: Turów in Poland and Jänschwalde and Boxberg in the Lausatia region of eastern Germany. The second one is in the industrial region of the Upper Silesia, Poland, where eleven (out of total twenty one) coal-fired power stations are located as well as many industrial and municipal sources of emission. The third one operates around the lignite power station Bełchatów in the central part of Poland. In 1999, the pattern of the SO_2 distribution in the country is similar; however, the concentration values are much lower. The changes in the concentration of SO_2 of the air are somewhat bigger (45%) than the range of the national emission changes reported. Three areas with raised concentrations can still be seen, but these regions are evidently much cleaner. This is due to a quite substantial (40%) reduction in the SO_2 emitted from Polish sources in 1999 compared to 1992 and due to accelerated reduction of emissions beyond Polish borders. From 1992 to 1999 sulphur emissions fell by 75% and by ca. 80% in Germany and in the Czech Republic, respectively (EMEP [7]). As the prevailing wind direction in central Europe is western, Poland has considerably benefited from these reduction of emissions, especially those from Germany. Both Jänschwalde and Boxberg power stations, being very close to the Polish border, were completely modernized, which reduced their emission by almost 90%. In the Turów power plant situated in the middle of the Black Triangle, which is still under modernization, the reduction of emission between 1992 and 1999 reached 75%. Together with the reduction of emission from the Czech sources these substantially improved the air quality in the Black Triangle Region.

Table 1

Comparison of the annual mean concentrations and total annual deposition of sulphur species predicted for 1992 and 1999.
Values averaged over (concentrations) or summarized for (deposition) the Polish area

Variable	Unit	1992	1999
Concentration of SO_2	$\mu g\ m^{-3}$	12.49	6.76
Concentration of SO_4^{2-}	$\mu g\ m^{-3}$	3.94	2.08
Total deposition	1000 t (S)/a	845.54	452.43

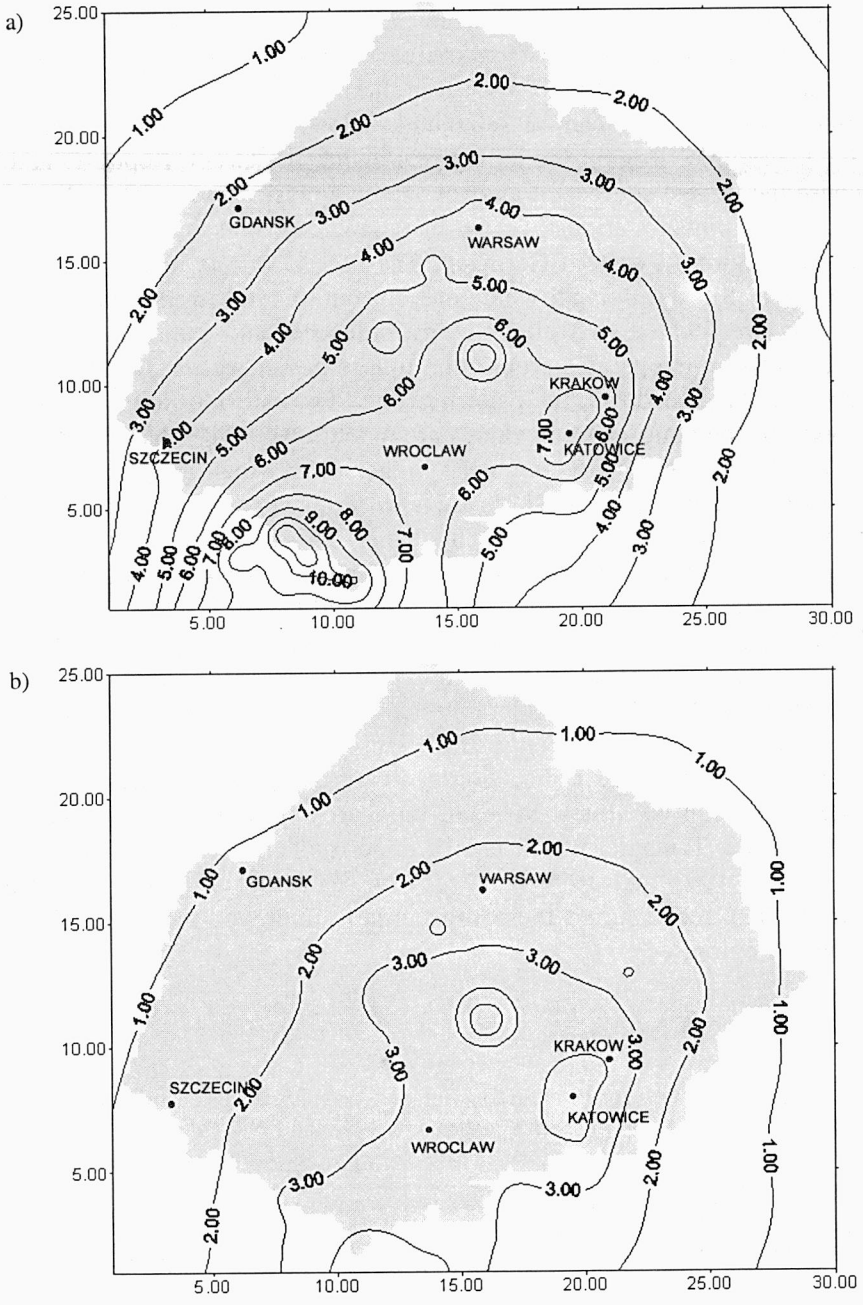

Fig. 2. Calculated yearly averaged SO_4^{2-} concentrations in Poland [μg (SO_4) m^{-3}] in 1992 (a) and in 1999 (b), JUDA-REZLER [16]

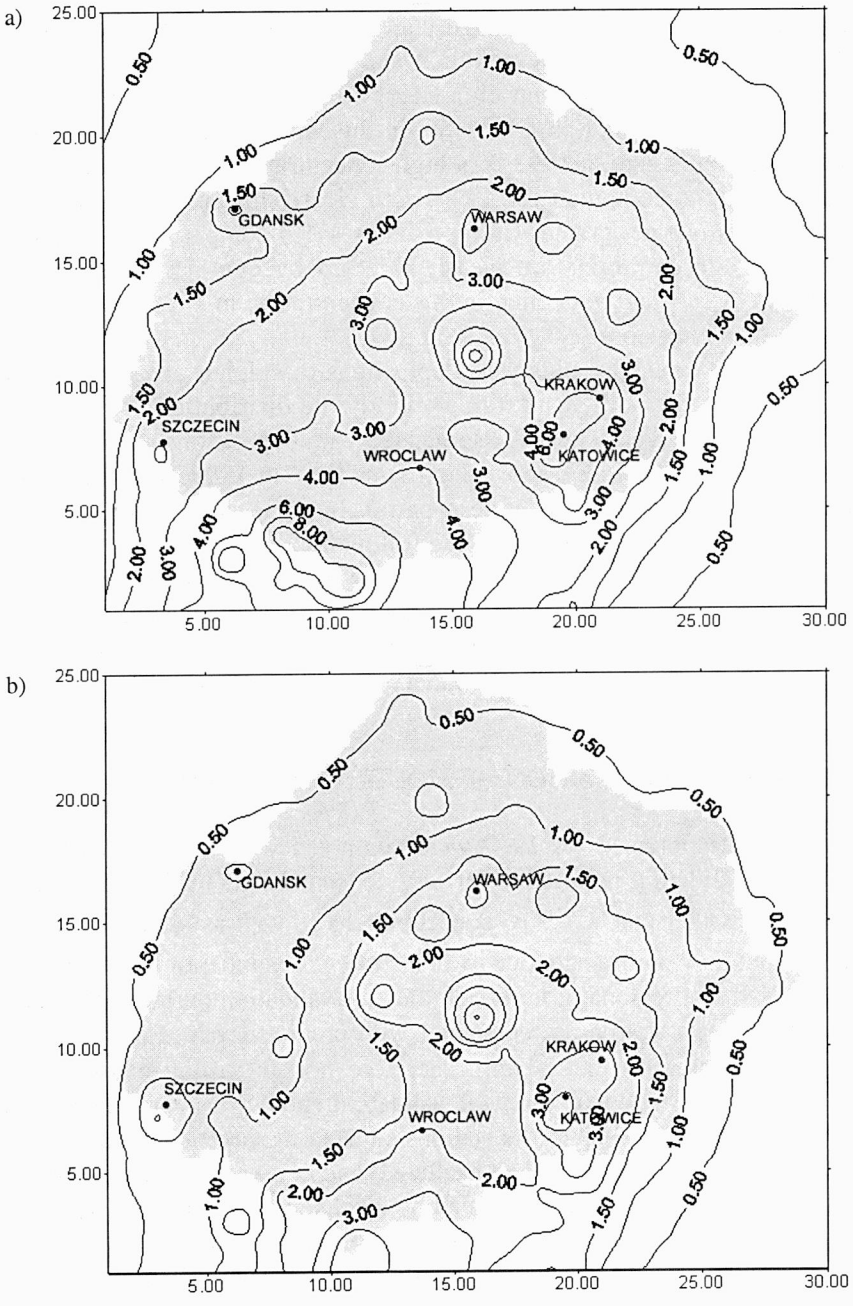

Fig. 3. Calculated annual total (dry + wet) sulphur deposition in Poland [g(S) m^{-2} yr^{-1}] in 1992 (a) and in 1999 (b), JUDA-REZLER [16]

The maps of calculated annual average concentrations of sulphate are shown in figure 2. As sulphate is a secondary pollutant and has a longer residence time compared to sulphur dioxide, the concentration fields computed are smoother. The influence of emission sources is not so noticeable in this case, with the exception of the Black Triangle Region, where in 1992 very high concentrations of sulphates occurred. Sulphate concentrations computed for 1999 were clearly lower. The decline of sulphate concentrations is even greater (it approaches 47%) compared to sulphur dioxide. The sulphate concentrations are clearly affected by emission from the neighbouring countries. A substantial decline in this concentration in 1999 is mainly due to reduction of German emission.

Calculated annual total (dry plus wet) depositions of sulphur are shown in figure 3. Deposition patterns for both years follow closely the distribution of SO_2 emission sources in the modelling area. There is a large area with depositions above 2 g (S) m^{-2} yr^{-1} in 1992, and with depositions above 1 g (S) m^{-2} yr^{-1} in 1999, which covers most of Poland (except its northern and eastern parts). In these areas, many maxima of scattered local deposition can be found in the following locations: Black Triangle and Upper Silesia regions as well as surrounding of the bigger power stations. However, in 1999, depositions were approximately by a factor of 2 lower than in 1992. The decline corresponds well with the decline of emissions, especially from power stations. The substantial reduction of transboundary deposition can be clearly seen.

4. THE EVALUATION OF MODEL PERFORMANCE

The model results obtained for 1999 were compared with measurements. At this moment complete validation is possible for SO_2 concentrations in the air being measured and predicted. Other results (SO_4^{2-} concentrations as well as total sulphur depositions) are compared only preliminary due to an insufficient number of measurements.

The POLSOX-II is a regional grid model, thus for validation purposes the values of point measurements at a station are compared with predicted values averaged for the grid cell area (in our case, 30 km × 30 km). Therefore, not all existing station data could be used for validation. Stations chosen for validation should be representative of the climatic conditions in the grid area (the stations situated in specific conditions, for example, sites of a high elevation should be excluded) and of an average air quality within the grid area (station should not be influenced by local sources). Furthermore, the usual requirement of temporal data completeness should be met as well as the requirement of a statistically sufficient number of stations covering the entire area of interest.

Two monitoring networks, i.e. the National Network of Basic Stations and the Monitoring Network of the Sanitary Inspectorate, are in operation in Poland (IEP [11]). All results were analysed in terms of their completeness and site representativeness. The 53 stations situated in all of the 16 voivodeships in Poland were chosen

for model validation. The method used for station selection is described by JUDA-REZLER [14].

Table 2

The statistical analysis of the predicted and measured annual mean SO_2 concentrations at monitoring stations in 1999 (a total number of stations considered $N = 53$). The statistical parameters are defined in the text

Parameter	Unit	Value
Measured mean	$\mu g\ m^{-3}$	7.93
Predicted mean	$\mu g\ m^{-3}$	7.83
Standard deviation of the measured values	$\mu g\ m^{-3}$	5.69
Standard deviation of the predicted values	$\mu g\ m^{-3}$	4.56
Bias	$\mu g\ m^{-3}$	0.10
Normalised mean bias (NMB)	%	1.31
Fractional bias (FB)	–	0.01
Noise to signal ratio (NSR)	–	0.41
NMSE	–	0.17
Ratio of standard deviations (F)	–	0.64
Fractional standard deviation (FSD)	–	0.43
Correlation coefficient (r)	–	0.82
Index of agreement (IA)	–	0.89
Explained variance (VWM)	–	0.68

In table 2, statistics for measured and predicted annual average SO_2 concentrations for chosen stations have been calculated. These include the number of stations N, spatial mean values, standard deviations and chosen statistical measures. The comprehensive package of 18 criteria for evaluating the performance of a model was proposed by the authoress (JUDA [12]). From that package the most significant criteria have been selected herein. Additionally, the widely used fractional bias as well as fractional standard deviation are included. The following measures are calculated:

• Measures of difference: mean bias (MB), normalized mean bias (NMB), fractional bias (FB) and noise to signal ratio (NSR). It should be pointed out that bias in this study is calculated as the difference between *measured* and *predicted* values, therefore a negative value of bias indicates that the model is overpredicting. The same notation is used elsewhere, e.g. by USEPA. In the same studies, however, the opposite notation is used (e.g. KARPPINEN et al. [18], BRANDT et al. [6]). Unbiased models will have small bias and a value of FB close to zero.

• Measures of model error: normalised mean square error (NMSE), previously named dimensionless MSE. Low value of NMSE indicates a less scattered model.

• Measures of standard deviations: ratio of standard deviations (F), fractional standard deviation (FSD). A model that gives a good estimation of the spread of the measurements will have a value of F close to unity and a value of FSD close to zero.

• Measures of correlation: correlation coefficient (r), index of agreement (IA). IA, which is both a relative and bounded measure, was proposed by WILLMOTT [23] as an alternative to r. A model that allows a perfect agreement between the measured and predicted values will have a value of r and IA equal to unity.

• Explained variance VWM which is a measure of how much of the observed variance is explained by the model. If the MSE is equal to zero, VWM is equal to unity. For an effective model, the VWM should be greater than 0.3 (30%).

Detailed explanation for the majority of these statistical parameters can be found in JUDA [12]. The fractional bias and fractional standard deviation are defined by:

$$FB = 2\frac{\overline{O} - \overline{P}}{\overline{O} + \overline{P}} \tag{1}$$

and

$$FSD = 2\frac{\sigma_0^2 - \sigma_p^2}{\sigma_o^2 + \sigma_p^2}, \tag{2}$$

where $\overline{O}$ and $\overline{P}$ are the mean values of observed and predicted series, respectively; σ_o and σ_p are the standard deviation of observations and predicted values, respectively.

The statistical results in table 2 testify to a fairly good agreement between measurements and calculations. The predicted mean concentration is very close to the measured mean, the relative bias is below 1.5%, the FB is close to zero, which means that the model is unbiased. The NMSE is small and equals 0.17. The estimation of the spread of the measurements is not very good, as a value of F equals 0.64 and a value of FSD equals 0.43. However, this could be partly explained by the structure of the model. The standard deviation of the predicted, as a grid average, values will always be smaller than the standard deviation of the point observations. This is especially true for the country, where substantial spatial gradients in concentrations occur (see figure 1) because of unequal emission distribution. The correlation measures (r, IA) exceeding 0.8 indicate very good correlation between measurements and predictions. Finally, the explained variance is equal to 0.68 which testifies to a good model performance.

In figure 4, the comparison of measured and predicted annually averaged SO_2 values are shown as scatter plots. The fractional biases (FB) were calculated for each

station. The FB values correspond with the ranges in the scatter plot: FB between –0.67 and 0.67 corresponds to the deviation in the scatter plot within a factor of two (upper and lower dashed lines in the scatter plot), and between –0.22 and 0.29 describes relative differences within 25%, i.e. stations with the best agreement. In the case of SO_2 concentrations, 85% of the station observations are within a factor of two, while for 30 out of 53 stations the difference is within +/– 25%. There are, however, a few stations, for which the discrepancies exceed a factor of two. That is especially the case of two stations situated on the Polish-German border, which are probably influenced by local emission sources and the values measured are much higher than the predicted ones. For the other six stations – which measured low concentrations – the predicted values are considerably overestimated. These stations are probably situated in the cleaner parts of the grids to which they belong and are not representative of the mean air quality there.

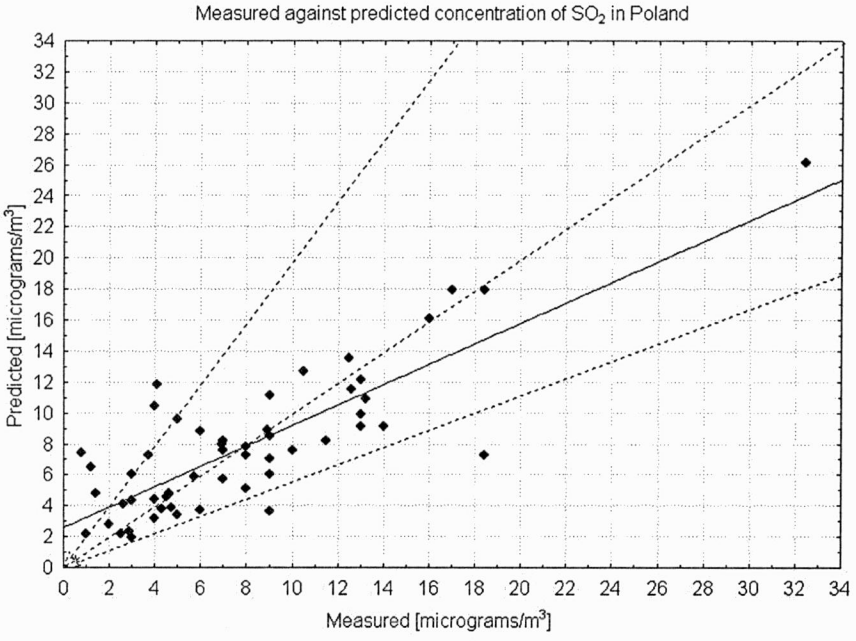

Fig. 4. Comparison of the yearly averaged measured and predicted SO_2 concentrations in Poland for 1999. Number of stations $N = 53$. Solid line represents the regression line; $y = 2.59 + 0.66\,x$. Dashed lines indicate perfect agreement (middle line) and difference by a factor of 2, JUDA-REZLER [16]

Sulphate in the air as well as sulphur deposition are measured in Poland only at four EMEP stations. One of them, however, has to be excluded from the comparison due to its location at a mountain top (Śnieżka, 1603 m asl). Comparison of the measured and predicted values for the remaining three stations is given in table 3 and table

4 for annual average sulphate concentrations and annual total sulphur depositions, respectively. For each station fractional bias was computed; however, the number of stations did not allow us to calculate any other statistical measures.

Table 3

Comparison of the predicted and measured annual mean SO_4^{2-} concentrations at EMEP monitoring stations in 1999. Measured data taken from EMEP (HJELLBREKKE [10])

Station name and EMEP number	Measured annual mean [$\mu g\ m^{-3}$]	Predicted annual mean [$\mu g\ m^{-3}$]	Fractional bias [/]
Jarczew, PL2	4.80	1.97	0.84
Łeba, PL4	3.75	0.93	1.21
Diabla Góra, PL5	2.13	0.89	0.82

The results given in table 3 indicate that the POLSOX-II model considerably underestimates sulphate concentrations in the air. The discrepancies exceed a factor of two for stations PL2 and PL5, and a factor of four for station PL4. The third station is, however, situated on the coast and thus its specific location and the presence of sea salt-containing sulphate is probably a reason of such a big difference between measured and predicted grid average values.

To some extent the model underestimations of sulphate concentrations might be due to the very simple scheme of SO_2 to SO_4^{2-} oxidation and due to rather simple dry deposition submodel.

Table 4

Comparison of the predicted and measured annual total sulphur deposition at EMEP monitoring stations in 1999. Measured data taken from IEP [11]

Station name and EMEP number	Measured annual total [$g(S)\ m^{-2}$]	Predicted annual total [$g(S)\ m^{-2}$]	Fractional bias [/]
Jarczew, PL2	1.01	1.12	–0.10
Łeba, PL4	0.78	0.51	0.42
Diabla Góra, PL5	0.59	0.57	0.03

Measured and predicted values of annual total (a sum of wet and dry) sulphur deposition are reported in table 4. Only wet deposition is measured directly, while dry deposition is estimated based on the measured concentration of sulphur species in the air. However, as other data are not available, the comparison between these data and predicted ones is presented. The results demonstrate that the POLSOX-II model predicts sulphur deposition quite satisfactorily – for stations PL2 and PL5 the agreement is very good, somewhat worst agreement was achieved again for station PL4.

The results of the model performance evaluation are encouraging, although complete model validation will be possible when a statistically sufficient set of measured data is available for both sulphate concentrations in the air and sulphur deposition. The results obtained so far show good model ability to predict long-term averages of SO_2 concentrations in the air and sulphur depositions for the area of Poland.

A poor agreement between predicted sulphate concentrations and measurements has five possible explanations: (1) chemical and physical description of the secondary particle formation and behaviour in the model is inadequate; (2) the quality of measurement is poor; (3) a part of measured sulphate is biogenic in origin; (4) sulphate concentrations are primary pollutants produced by combustion in small boilerhouses and ceramic domestic stoves; (5) a part of the measured sulphate is a secondary pollutant transported from the outside of the modelling area. The first explanation, however, is not consistent with a good agreement found between measured and predicted SO_2 concentrations and sulphur depositions and the mass conservation principle. The quality of the 1999 EMEP measurements was assessed, in which laboratory performance of all Polish stations (concerning measurements of sulphates in the air) was characterized as "very good" (EMEP [7]). The remaining points i.e. (3), (4) and (5), provide possible explanations of the discrepancies obtained. Option (3) is possible, as no biogenic emissions are considered in the total emissions. Option (4) is also possible as in the model sulphate acts almost exclusively as a secondary pollutant, only 5% of the total primary SO_x emissions is assigned to it. However, in author's opinion, the fifth possibility is most probable. As sulphate has a longer residence time compared to sulphur dioxide, it can be transported for longer distances. It appears, therefore, that the computational grid is big enough to include the most important sulphur dioxide sources from outside of Poland, but is not big enough to include all sources, which are responsible for sulphate inflow to Poland. Increased efforts should be dedicated to further investigation of this concept. Introduction of non-zero boundary conditions for sulphate might probably be one of the possible solutions.

5. CONCLUSIONS

The performance and evaluation of the POLSOX-II numerical two-dimensional Eulerian grid model applied to the area of Central Europe have been presented. Model results – annual averages of sulphur species as well as total annual sulphur deposition – have been compared for two selected years, representing past and current air quality in Poland. The results show a substantial improvement of the air quality in the country, with the changes somewhat greater than the range of the reductions of national emission reported. This is due to the accelerated reduction of emissions beyond Polish borders, especially those in Germany. However, while the calculated values are cur-

rently approximately by a factor of 2 smaller than those at the beginning of the nineties, certain areas still experience elevated levels of SO_x concentrations and sulphur depositions.

The model estimates were verified by measurements. Very good agreement was obtained between measured and calculated sulphur dioxide concentrations for which a package of statistical parameters have been used. Calculated sulphate concentrations as well as sulphur deposition have been compared only preliminary due to the lack of a sufficient number of measurements. The results of the model performance evaluation prove that the model allows an accurate prediction of the long-term averages of sulphur dioxide concentrations in the air and total sulphur depositions for the area of Poland. However, the sulphate concentrations predicted based on the model are substantially underestimated. It was suggested that the computational grid is not big enough to include all sources, which are responsible for sulphate inflow to Poland. Consequently, it seems that the European background concentrations should be added to the sulphate concentrations calculated with POLSOX-II.

The results presented herein are encouraging to apply simple models based on the available input data of known quality as well as on crude but well-characterised parameterisations of physical and chemical processes for assessing long-term air quality on a regional scale.

ACKNOWLEDGEMENTS

The authoress acknowledges Dr. Aleksander Warchałowski at Warsaw University of Technology for providing detailed emission data for Poland. Doctor Ian Lowles, Westlakes Research Institute, UK, is acknowledged for useful comments to the manuscript.

REFERENCES

[1] ABERT K., BUDZIŃSKI K., JUDA-REZLER K., *Regional air pollution models for Poland*, Ecological Engineering, 1994a, 3, 225–244.

[2] ABERT K., BUDZIŃSKI K., JUDA-REZLER K., *Influence of the meteorological fields on the annual concentration and deposition of sulphur and nitrogen compounds*, Polish Contribution-in-kind to EMEP, 1994b, EMEP/Poland - Rep. 1/94, Warsaw University of Technology, Warsaw, Poland, 32 pp.

[3] BARTNICKI J., *A simple filtering procedure for removing negative values from numerical solution of the advection equation*, Environmental Software, 1989, 4, 187–207.

[4] BARTNICKI J., *An Eulerian model for the long range transport of heavy metals over Europe: model description and preliminary results*, Water, Air and Soil Pollution, 1994, 75, 227–235.

[5] BARTNICKI J., HREHORUK J., GRZYBOWSKA A., MAZUR A., *Regional model for atmospheric transport of heavy metals over Poland*, [in:] P. Anttila, J. Kamari and M. Tolvanen (Eds.), *Proceedings of the 10th Air Clean World Congress*, Espoo, Finland, 1995, pp. 339–350.

[6] BRANDT J., CHRISTENSEN J.H., FROHN L.M., PALMGREN F., BERKOWICZ R., ZLATEV Z., *Operational air pollution forecasts from European to local scale*, Atmospheric Environment, 2001, 35 (Suppl. 1), S91–S98.

[7] EMEP, *Transboundary acidification, eutrophication and ground level ozone in Europe – Summary Report 2001*, EMEP-W Report 1/2001, Oslo, 2001, Norway.

[8] EMEP, *Data; Detailed reports per country; Poland; Emissions split by source sector*, Internet, EMEP webside, 2003, http://www.emep.int/areas/ sectortab/PL_SO2.html.

[9] GOLDER D., *Relations among stability parameters in the surface layer*, Boundary Layer Meteorology, 1972, 3, 47–58.

[10] HJELLBREKKE A.-G., *Data report 1999. Acidifying and eutrophying compounds.* Part 1. *Annual summaries*, EMEP/CCC-Report 2/2001, Norwegian Institute for Air Research, Kjeller, Norway.

[11] IEP, 2001. *Air pollution in Poland in 1998–1999* (in Polish), Report of the Inspectorate of Environmental Protection, Warsaw, Poland, 311 pp.

[12] JUDA K., *Modelling of the air pollution in the Cracow area*, Atmospheric Environment, 1986, 20, 2449–2558.

[13] JUDA-REZLER K., *Risk assessment of airborne sulphur species in Poland*, [in:] Preprints of the 26th NATO/CCMS International Technical Meeting on Air Pollution Modelling and its Application, Istanbul Technical University, Istanbul, Turkey, 26–30 May, 2003, pp. 10–18.

[14] JUDA-REZLER K., *Current sulphur dioxide concentration levels in Poland – model estimates and comparison to observations*, [in:] L. Pawłowski, M.R. Dudzińska and A. Pawłowski (Eds), *Environmental Engineering Studies. Polish Research on the Way to the EU*, Plenum/Kluwer, New York, 2003b, pp. 13–22.

[15] JUDA-REZLER K., ABERT K., *Description of the POLSOX-I model. Results from the POLSOX-I model*, [in:] H.M Apsimon (Ed.), *Emission Abatement Strategies and the Environment (EASE) – Final Report*, Imperial College of Science, Technology and Medicine, London, 1997, pp. 381–398.

[16] JUDA-REZLER K., *Universal goal function for the Integrated Assessment Models for air pollution* (in Polish), Scientific Works of Warsaw University of Technology, Environmental Engineering, Publishing House of the Warsaw University of Technology, 2004, Vol. 41, p. 131.

[17] KAMIŃSKI J.W., PLUMMER D.A., NEARY L., MCCONNELL J.C., STRUZEWSKA J., ŁOBOCKI L., *First application of MC2-AQ to multiscale air quality modelling over Europe*, Physics and Chemistry of the Earth, 2002, 27, 1517–1524.

[18] KARPPINEN A., KUKKONEN J., ELOLÄHDE T., KONTTINEN M., KOSKENTALO T., RANTAKRANS E., *A modelling system for predicting urban air pollution: model description and applications in the Helsinki metropolitan area*, Atmospheric Environment, 2000, 34, 3723–3733.

[19] LITYŃSKA Z., *Determination of the mixing layer height and wind shear for the Pasquill stability classes using data from four radio-sounding and connected ground-level meteorological stations in Poland* (in Polish), PR 8 Interim Report, Warsaw University of Technology, Warsaw, 1985, Poland, 25 pp.

[20] LOWLES I., APSIMON H., JUDA-REZLER K., ABERT K., BRECHLER J., HOLPUCH J., GROSSINHO A., *Integrated assessment models – tools for developing emission abatement strategies for the Black Triangle region*, Journal of Hazardous Material, 1998, 61, 229–237.

[21] MARKIEWICZ M., *The Gaussian air pollution dispersion model with variability of the input parameters taken into account*, Environment Protection Engineering, 1994, 19 (1–4), 123–141.

[22] ULIASZ M., OLENDRZYNSKI K., BARTNICKI J., *Mesoscale modelling of transport and deposition of heavy metals in southern Poland*, [in:] S.-E. Gryning and N. Chaumerliac (Eds), *Air Pollution Modelling and Its Application XII*, Plenum Press, New York and London, 1998, pp. 53–61.

[23] WILLMOTT C.J., *Some comments on the evaluation of model performance*, Bull. Am. Met. Soc., 1982, 63, 1309–1313.

[24] YANENKO N.N., *The method of fractional steps*, Springer-Verlag, New York, 1971.
[25] ZIENKIEWICZ O.C., TAYLOR R.L, *The Finite Element Method*, Fourth Edition, McGraw-Hill, New York, 1989.

MODELOWANIE ROZPRZESTRZENIANIA SIĘ ZWIĄZKÓW SIARKI NA OBSZARZE POLSKI

Polska jest krajem posiadającym duże pokłady węgla kamiennego i brunatnego, co powoduje, że choć udział procentowy węgla w strukturze zużycia nośników energii zmalał w ciągu ostatniego dwudziestolecia o około 15%, to energia pierwotna w ponad 60% jest nadal pozyskiwana z paliw stałych. Mimo znacznych redukcji emisji SO_2 w ostatnich latach, Polska nadal należy do największych emiterów tego związku w Europie, co stanowi potencjalne zagrożenie dla środowiska i zdrowia ludzkiego. Aby ocenić to zagrożenie, opracowano zintegrowany system modelowania ROSE (Risk of airborne sulphur species to the environment), w którym zastosowano koncepcję *jedno zanieczyszczenie – wielość efektów*, co pozwala uwzględnić i ocenić wszystkie negatywne skutki oddziaływania związków siarki na środowisko. Jedną z części składowych systemu ROSE jest regionalny, numeryczny model rozprzestrzeniania się związków siarki w atmosferze, POLSOX-II, który zaprezentowano w pracy. Przeprowadzono obliczenia dla dwóch wybranych lat: 1992 i 1999, reprezentujących odpowiednio stan zanieczyszczenia atmosfery w początkowym okresie transformacji ustrojowej oraz stan aktualny. Wyniki modelowania zweryfikowano, porównując je z danymi pomiarowymi. Porównanie przeprowadzono dla średniorocznych stężeń SO_2 i SO_4^{2-} oraz całkowitej rocznej depozycji siarki. Wyniki pokazują, że zanieczyszczenie powietrza i ekosystemów w Polsce przez związki siarki znacznie zmalało w analizowanym okresie, lecz w niektórych regionach jest ono nadal duże. Dla stężeń SO_2 przeprowadzono kompleksową weryfikację wyników modelowania, korzystając z danych pomiarowych uzyskanych z 53 stacji monitoringu zanieczyszczeń powietrza. Uzyskano bardzo dobrą zgodność wyników modelowania z wartościami pomiarowymi. Dostępne dane pomiarowe wskazują ponadto, że otrzymane wyniki obliczeń depozycji siarki na podłożu są dobrze zbieżne z wynikami pomiarów, niezadowalająca jest natomiast jakość predykcji modelu w odniesieniu do stężeń SO_4^{2-}. Omówiono ten problem.