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ESTIMATION OF THE EXCHANGE OF SULPHUR POLLUTION OVER THE BALKAN REGION IN 1995-2000

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ABSTRACT

The Bulgarian dispersion model EMAP is used to estimate the sulphur pollution over the Balkan region for the period 1995-2000. A sub-domain of the EMEP grid is chosen containing 12 countries. As meteorological driver the operational DWD "Europa-Model" is used. The source input is the official EMEP emission data. Monthly calculations are made having the last moment fields from the previous month as initial conditions for the next. The boundary conditions are set to zero so the influence of the other European sources is not accounted for. According to the EMEP methodology multiple runs are made setting every time the sources of various countries to zero. The impact of every country in the pollution of all others is estimated.

Key Words: Dispersion Modelling, Sulphur Dioxide, Blame matrix, Multi-year estimates.

1. INTRODUCTION

The Bulgarian dispersion model EMAP is used to estimate the concentration in air and in precipitation water and the total deposition of oxidized sulphur over southeast Europe for the six-year period 1995 - 2000 due to sources from 12 countries (Table 1). As only sources from these countries are handled, the results can be considered as an estimate of their impact on the acid pollution of the region as well as an estimate of the reciprocal pollution.

Table 1. List of countries and their notations.

Albania	Bosnia and Herzegovina	Bulgaria	Croatia	Serbia and Montenegro	The FYR Macedonia
AL	BH	BG	HR	YU	MK
Moldavia	Romania	Turkey[1]	Slovenia	Greece	Hungary
MO	RO	TR	SL	GR	HU

(1) The part inside the model domain.

2. SHORT DESCRIPTION OF THE EMAP MODEL

EMAP (Syrakov, 1995) is a simulation model that allows describing the dispersion of multiple pollutants. The processes of horizontal and vertical advection, horizontal

and vertical diffusion, dry deposition, wet removal, gravitational settling (aerosol version) and the simplest chemical transformation (sulphur version) (Seinfeld, 1986) are accounted for in the model. Within EMAP, the semi-empirical diffusion-advection equation for scalar quantities is treated. The governing equations are solved in terrain-following coordinates. Non-equidistant grid spacing is settled in vertical directions. The numerical solution is based on discretization applied on staggered grids using the splitting approach. Conservative properties are fully preserved within the discrete model equations. Advective terms are treated with the TRAP scheme, which is a Bott-type one. Displaying the same simulation properties as the Bott scheme (explicit, conservative, positive definite, transportability, and limited numerical dispersion), the TRAP scheme proves to be several times faster (Syrakov, 1996; Syrakov and Galperin, 1997). The advective boundary conditions are zero at income flows and "open boundary" - at outcome ones. Turbulent diffusion equations are digitized by means of the simplest schemes – explicit in horizontal, and implicit in vertical direction. The bottom boundary condition for the vertical diffusion equation is the dry deposition flux, the top boundary condition is optionally "open boundary" and "hard lid" type. The lateral boundary conditions for diffusion are "open boundary" type. In the surface layer (SL), a parameterization is applied permitting to have the first computational level at the top of SL. It provides a good estimate for the roughness level concentration and accounts also for the action of continuous sources on the earth surface (Syrakov and Yordanov 1996). A similarity theory based PBL model (Syrakov and Yordanov 1997) is built in the model producing 3D velocity and turbulence fields on the base of minimum meteorological information – the wind and temperature at geostrophic level and the surface temperature.
The model is evaluated and validated during the ETEX-II intercalibration study - ranged 9th among 34 models (Syrakov and Prodanova, 1998). It is validated on the database of 1996 EMEP/MSC-E intercalibration of heavy metal models (Syrakov and Galperin, 1997).

3. MODEL DOMAIN, PARAMETERIZATION AND INPUT DATA

The aim of this modeling is to estimate the sulphur pollution in the region of Southeast Europe, taking a territory of 38×28 EMEP 50×50 km^2 grid cells with Bulgaria in the center (see figure 1). Every cell is divided to four 25×25 km^2 cells. The chosen territory includes entirely all 11 countries of interest and partly other territories. In the created versions of EMAP, a 5-layer vertical structure is used. The first four layers have representative levels at 50, 200, 650 and 1450 m with layer boundaries 20-100, 100-375, 375-995, 995-1930 m. The 5th layer accounts parametrically for the free atmosphere. The volume of this layer is so big that the concentration tends to be zero there, although it can contain some mass.
Two kinds of input are necessary for EMAP performance: sources and meteorological data.

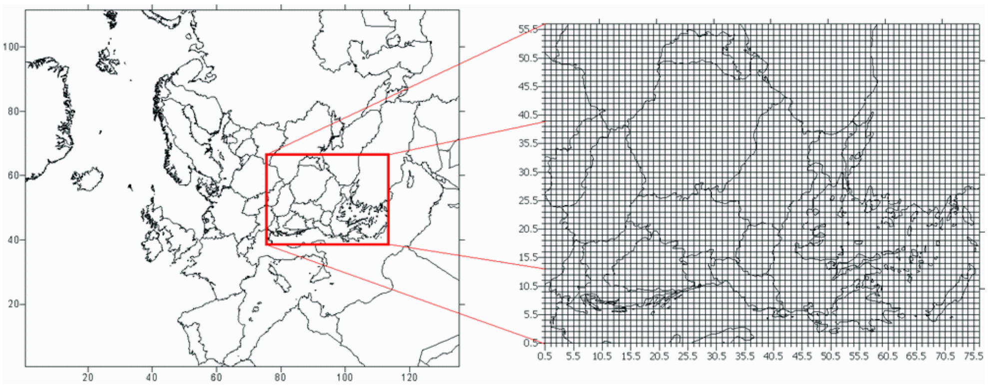

Figure 1. Nesting the model domain in the EMEP-domain

3.1 EMISSIONS

The sources (stationary, monodisperse - SO_2 only) are determined through an emission inventory based on the CORINAIR methodology. They correspond to the official 50x50 km^2 data reported by the corresponding governmental authorities to EMEP's MSC-West and can be downloaded from its web site (http://www.emep.int/). Additional mass-conservative redistribution of these data is made over the finer grid of 25-km space resolution. The emissions are provided in mass units per second. All sources are divided in two classes: high sources (like high and very strength industrial stacks etc.) called Large Point Sources (LPS) and area sources (AS) –the sum of all low and diffusive sources in the given grid cell. As all LPS are supplied with high stacks, the emission of these sources is prescribed to be released in layer 2, i.e., between 100 and 375 m.. In this paper the present scheme of EMEP has been applied. In it the sources are divided in 11 SNAP (Selected Nomenclature for reporting of Air Pollutants) sectors and the data gaps (in former schemes) are filled. The study of the contribution of the each sector is not a subject of this research, therefore each sector is treated as AS or LPS, according to table 3. The annual emission trend is shown on table 2. According the inventory the number and the strength of the sources are changed in the years. It can be seen that by the most of the countries the emitted quantities with a few exceptions decreased during the period of the study. By 9 countries (Albania, Bulgaria, Bosna&Herzegovina, Greece, Hungary, Croatia, Moldova, Slovenia and Serbia&Montenegro) these quantities for 2000 are smaller then for 1995, by 2 countries (Macedonia and Hungary) there is no significant change and only by Turkey the emitted mass for 2000 is greater than for 1995. Generally, the total released mass of sulphur dioxide decreased from 55456 10^2t to 46531 10^2t., i.e. with 16%. This rate is much smaller in comparison with other regions of Europe for the same period (Lövblad et al., (Eds.) 2004)

Table 2. Annual trend of the emission strength (unit: 100t/year). The boxes where the values are greater then the values for the previous year are shown in orange.

year	source	AL	BG	BH	GR	HR	HU	MK	MO	RO	SL	TR	YU	all countr.
1995	ARS	290.7	2334.9	1776.0	1303.2	452.8	2587.0	388.5	14.4	2758.1	197.3	2021.8	670.0	14794.8
	LPS	429.3	12425.1	3024.0	3976.8	251.2	4462.6	661.5	626.2	6361.9	1052.7	3439.9	3950.0	40661.1
	Total	720.0	14760.0	4800.0	5280.0	704.0	7049.6	1050.0	640.6	9120.0	1250.0	5461.7	4620.0	55455.9
1996	ARS	290.7	2242.5	1776.0	1404.9	449.2	2360.0	388.5	13.0	2758.1	152.4	2201.8	520.0	14557.3
	LPS	429.3	11957.5	3024.0	3775.1	212.8	4372.3	661.5	657.3	6361.9	967.6	3746.1	3820.0	39985.3
	Total	720.0	14200.0	4800.0	5180.0	662.0	6732.3	1050.0	670.3	9120.0	1120.0	5947.9	4340.0	54542.5
1997	ARS	290.7	2216.3	1776.0	1384.0	446.3	1947.7	388.5	8.7	2758.1	138.9	2271.0	580.0	14206.3
	LPS	429.3	11433.7	3024.0	3726.0	357.7	4637.4	661.5	352.6	6361.9	1041.1	3863.9	4640.0	40529.0
	Total	720.0	13650.0	4800.0	5110.0	804.0	6585.1	1050.0	361.3	9120.0	1180.0	6135.0	5220.0	54735.4
1998	ARS	290.7	2192.0	1776.0	1512.8	424.1	1290.9	388.5	7.2	2758.1	109.2	2417.4	690.0	13857.1
	LPS	429.3	10318.0	3024.0	3667.2	470.9	4627.0	661.5	313.6	6361.9	1120.8	4113.0	4520.0	39627.1
	Total	720.0	12510.0	4800.0	5180.0	895.0	5917.9	1050.0	320.8	9120.0	1230.0	6530.4	5210.0	53484.1
1999	ARS	355.3	618.4	342.3	1020.9	446.7	865.1	205.7	48.7	916.4	138.5	2545.7	633.0	8136.6
	LPS	214.7	8811.6	3727.7	4379.1	463.3	5034.9	844.3	71.3	8203.6	901.5	3940.5	2917.0	39509.5
	Total	570.0	9430.0	4070.0	5400.0	910.0	5900.0	1050.0	120.0	9120.0	1040.0	6486.2	3550.0	47646.2
2000	ARS	361.6	644.0	352.4	913.1	284.7	712.6	205.7	52.7	916.4	131.9	2555.3	690.1	7820.4
	LPS	218.4	9176.0	3837.6	3916.9	295.3	4147.4	844.3	77.3	8203.6	858.1	3955.5	3179.9	38710.4
	Total	580.0	9820.0	4190.0	4830.0	580.0	4860.0	1050.0	130.0	9120.0	990.0	6510.8	3870.0	46530.8

Table 3. SNAP Sectors and their description

SNAP	Description	assumed
S1	Combustion in energy and transformation industries (stationary sources)	LPS
S2	Non-industrial combustion plants (stationary sources)	AS
S3	Combustion in manufacturing industry (stationary sources)	AS
S4	Production processes (stationary sources)	AS
S5	Extraction and distribution of fossil fuels and geothermal energy	AS
S6	Solvent use and other product use	AS
S7	Road transport	AS
S8	Other mobile sources and machinery	AS
S9	Waste treatment and disposal	AS
S10	Agriculture	AS
S11	Other sources and sinks	Not incl.

The monthly emissions are obtained using the following dimensionless annual variation coefficients, recommended by MSC-E (Table 4):

Table 4. Annual variation coefficients

J	F	M	A	M	J	J	A	S	O	N	D
1.34	1.30	1.18	1.00	0.73	0.69	0.69	0.73	0.83	1.00	1.17	1.34

3.2 METEOROLOGICAL DATA

An important advantage of the used model is that, due to the built in PBL model, it utilizes only numerical analysis and forecast data from the world weather centers, distributed via the Global Telecommunication System of the World Meteorological Organization. For this task the meteorology input has a time resolution of 6 hours. It consists of the sequence of analyzed U_{850}, V_{850}, T_{850} and T_{surf} fields and 6-hour forecast for precipitation from the standard 50×50 km^2 output of the former "Europa-Model" of the German Met. Service (DWD). On this base, the PBL model calculates U-, V-, W- and K_z-profiles at each grid point. It provides also u_* and SL universal profiles necessary in SL parameterization. The roughness and the Coriolis parameter fields are pre-set additional input to the PBL model. Orography height, surface type (sea-land mask) and roughness height are to be provided for each grid location. Initial concentration field is optionally introduced (spin-up fields).

3.3 SULPHUR PARAMETRISATION AND OTHER MODEL PARAMETERS

Two species of sulphur in the air are considered: gaseous sulphur dioxide SO_2 and particulate sulfate $SO_4^{=}$. Sources emit SO_2 only, in the air it is transformed to sulfate. The pollutant specific model parameters used are given in Table 4.

Table 5. Specific model parameters

Pollutant	SO_2		$SO_4^{=}$	
Transformation rate constant α_{tr} [h^{-1}]	0.01 (winter) - 0.04 (summer)			
Wet removal constant γ [mm^{-1}],	0.3		0.2	
surface type	sea	land	sea	land
Dry deposition velocity V_d [m/s]	0.01	0.03	0.002	0.006

The other model parameters are: horizontal diffusion coefficients Kx= Ky= 5.10^4 m^2/s and time step Du= 0.25 h.

4. CALCULATION RESULTS

Monthly runs with the above mentioned sources are performed. The output consists of the following fields: monthly dry (DD), wet (WD) and total (TD) depositions, monthly concentration mean in air (CA) and, from the meteorological driver, monthly sum of the precipitation (SP). Additionally is calculated the concentration in the precipitation (CP) for each month. Then, the mean annual value of each of the above mentioned fields $\overline{p}$ is calculated according to the formula:

$$\overline{p} = \frac{\sum_1^n m_i p_i}{\sum_1^n m_i}, \; i = 1, n, \qquad (1)$$

where m_i is the duration of the month i, and p_i is the corresponding monthly value of the parameter. Finally, using (1) again, where m_i is the duration of each year 1995 - 2000, and p_i is the corresponding mean annual value, the average of the concentration in air, concentration in precipitation water and the total deposition in the whole period is obtained. The spatial distribution of these parameters is shown on figure 2, figure 3 and figure 4.

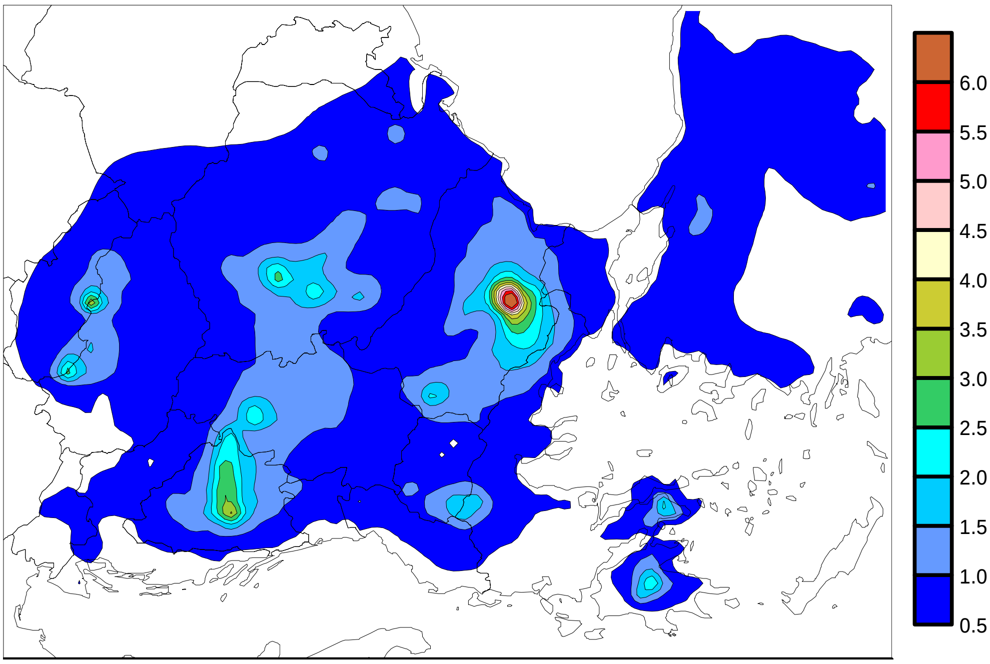

Figure 2. Mean annual concentration in air (unit: μg (S)/m^3) of oxidized sulphur in 1995-2000.

In figure 2, it is shown that in the main part of the model domain the concentration in surface air is below 1 μg/m^3; over a part of Northern Greece, Bulgaria, Romania, Serbia and Montenegro and Hungary it is between 1 and 2 μg/m^3 and only over small areas in Bosnia and Herzegovina, Romania, northern Hungary, in Bulgaria (the region of Sofia) and in Greece (Athens) it is over 3 μg/m^3. The most polluted region is that of Southeast Bulgaria, the place around the most powerful source in the region - "Maritsa Iztok" Thermal Power Plant. This TPP is a set of 3 neighboring coal-burning plants. They are so close to each other that they occupy a single 25-km cell. In its vicinity the total the concentration reaches 7 μg/m^3.

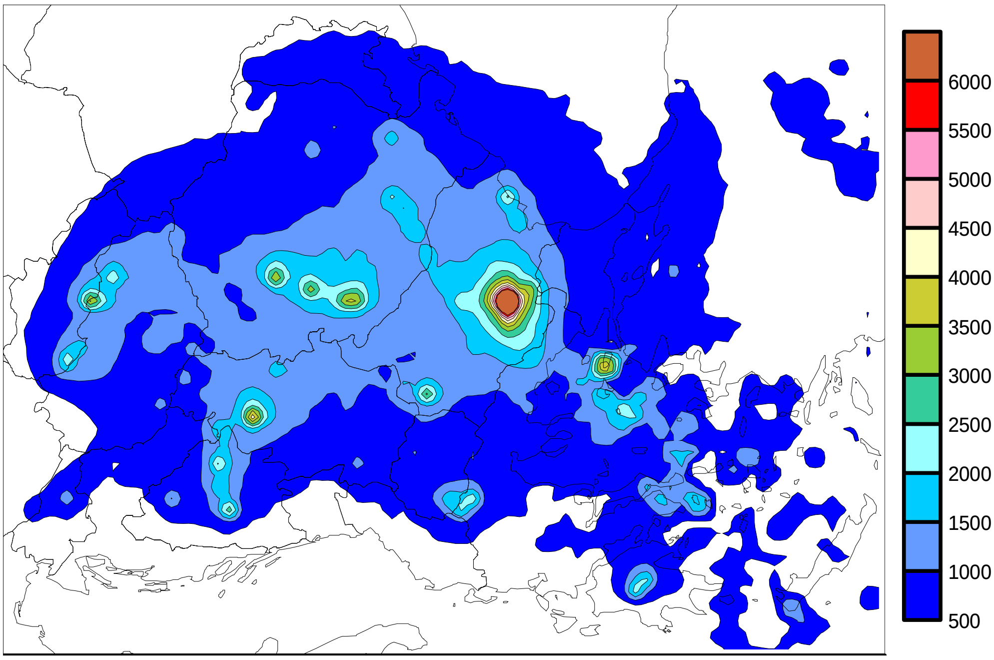

Figure 3. Mean annual total deposition (unit: mg (S)/m^2) of oxidized sulphur in 1995-2000.

The shape of isopleths of the mean annual total deposition is similar. Over the main part of the model domain it is below 2 g/m^2 only over small areas ("hot spots") in Bosnia and Herzegovina, Bulgaria, Romania and Hungary it is between 3 and 4 g/m^2. Again, the most polluted region is that of Southeast Bulgaria, the place around "Maritsa Iztok" TPP, where the deposition reaches 13.5 g/m^2.

The picture of the distribution of the concentration in the precipitation is more irregular then the others. In the main part of the model domain it is below 2 mg/l and over a few "hot spots", mainly near the strength sources, it is between 2 and 5 mg/l. In spite of the irregular field of the precipitation, the highest concentration is again in the region of "Maritsa Iztok" TPP where it reaches 13.6 mg/l.

In Table 6, the time-averaged deposition budget matrix for oxidized sulphur is presented. It is obtained from the annual matrixes with the above-mentioned procedure. Every-year matrix is calculated after multiplication of the total deposition field to array containing the distribution of the territory of each country in the model domain. The deposition budget matrix shows the impact of each country to the sulphur pollution of the other countries. The last three rows show respectively the medium values of the total deposited quantity due to the sources from the country in the column header, the total emitted sulphur for this country and the percentage of deposited quantities from the yearly emitted ones. The last value can be treated as the relative part of the sulphur that remained in the domain. It can be noticed that the main part of the sulphur pollution emitted by each country is deposited over the country itself. It can be seen also that the percentage of the total deposited quantity is between 19% and 30%, the rest goes out of the model domain.

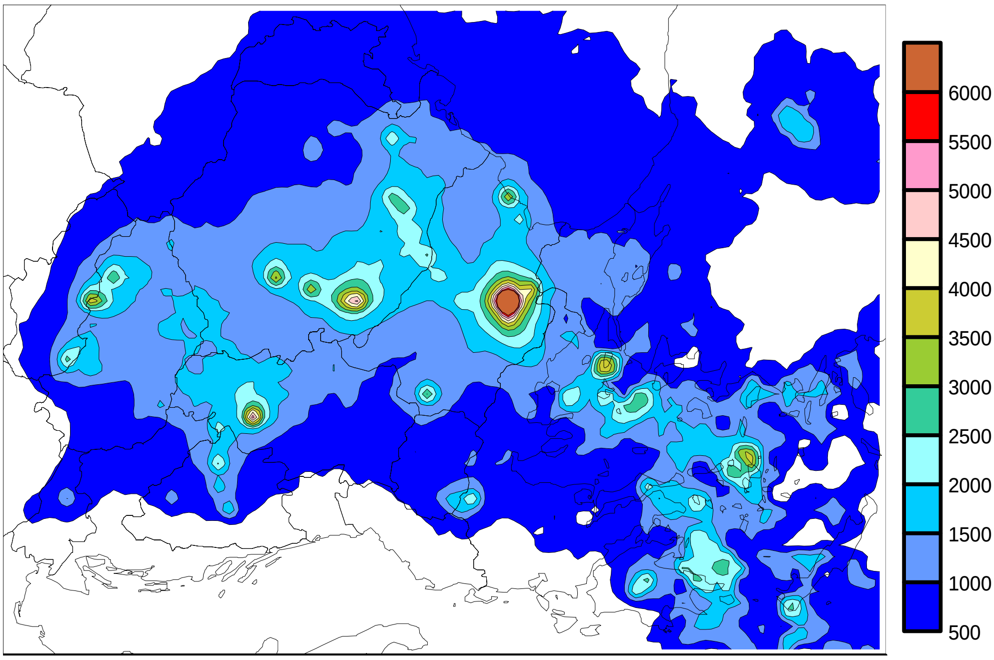

Figure 4. Mean annual concentration in the precipitation (unit: µg (S)/l) of oxidized sulphur in 1995-2000.

Table 6. Deposition budget matrix (unit 100 t). The shaded elements show the deposition quantity for each country due to its own sources.

emitter / receiver	AL	BG	BH	GR	HR	HU	MK	MO	RO	SL	TR	YU
AL	67.03	18.66	11.52	42.55	0.87	3.82	29.71	0.09	4.62	0.70	1.13	18.96
BG	8.30	1582.90	35.04	56.36	2.92	36.73	34.31	3.15	313.58	3.71	21.83	100.50
BH	3.43	11.13	483.66	4.88	22.66	41.75	2.80	0.13	11.21	10.69	0.48	67.50
GR	20.38	254.72	13.24	443.69	1.06	7.70	34.20	0.79	28.93	1.16	20.52	20.18
HR	1.30	7.80	102.95	2.58	70.90	44.54	1.26	0.09	8.98	33.47	0.34	26.98
HU	1.61	21.33	91.05	3.73	27.33	476.05	2.77	0.37	53.84	31.52	0.73	70.78
MK	17.93	58.67	7.55	65.18	0.53	4.96	77.65	0.14	9.41	0.54	1.81	20.70
MO	0.49	33.24	6.33	2.30	0.72	13.37	1.06	21.52	87.40	1.03	2.23	9.98
RO	8.32	392.07	152.43	26.77	15.45	274.30	19.22	17.07	1544.81	17.66	14.05	278.92
SL	0.18	1.99	8.20	0.56	15.80	9.48	0.22	0.03	2.51	68.05	0.09	4.89
TR	4.50	279.57	10.70	130.37	1.00	11.59	8.44	1.99	65.14	1.53	1037.56	18.72
YU	23.84	117.56	226.53	31.05	13.56	98.64	39.22	0.62	84.07	9.57	2.76	509.05
total dep.	198.42	3627.10	1346.59	1300.49	224.44	1651.32	293.86	74.79	2717.93	274.14	1347.22	1331.71
total em.	671.65	12394.65	4576.59	5163.19	759.04	6173.81	1050.00	373.86	9120.00	1134.93	6178.71	4468.00
%	29.55	29.04	29.31	25.16	29.56	26.65	27.99	19.69	29.80	24.16	21.82	29.89

The minimum of this value is for Moldova - the possible explanation is that this territory is close to the east border of the model domain and the main tropospheric transport is west east in these latitudes.

In the figure 5, the diagram of the annual trend of the deposition over each country due to the activity of all sources in the domain is shown. Generally, the trend is descending - for all countries, except Turkey, the deposed quantities for the last year are smaller then for the first year. Especially significant is the abatement in the deposition over the most polluted countries - Romania and Bulgaria. The ascending trend in Turkey can be explained with the increasing of the emissions there.

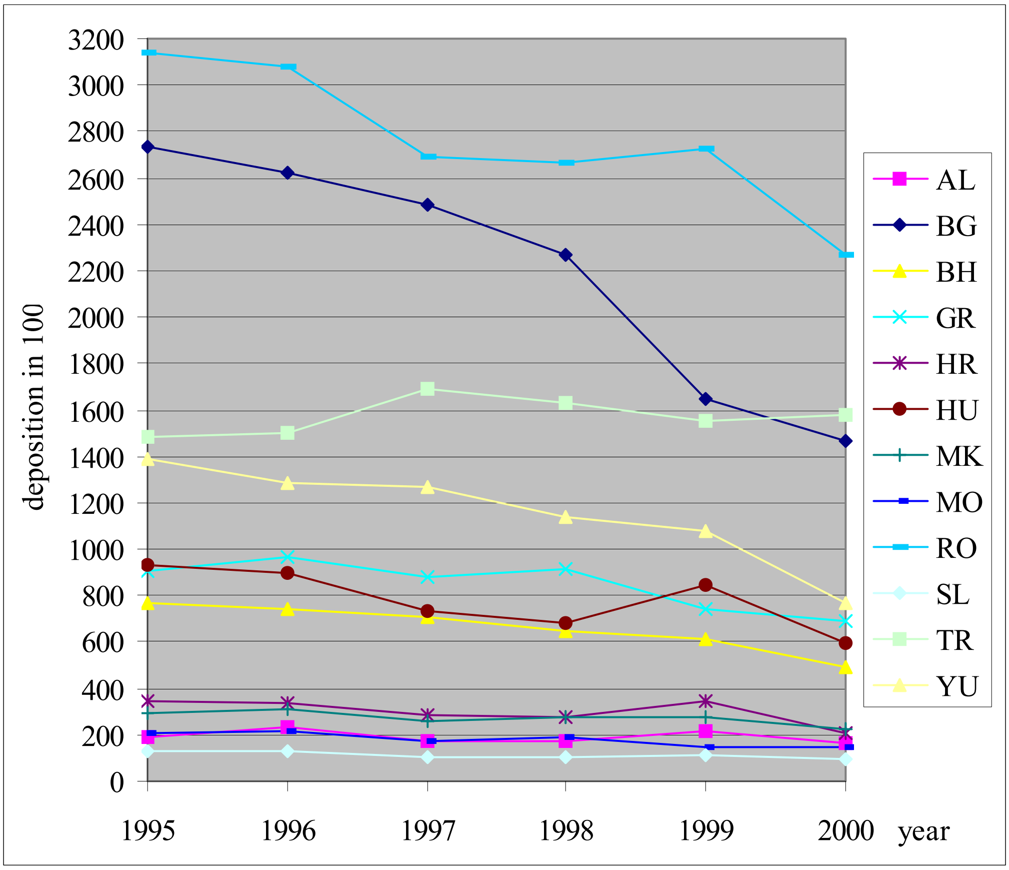

Figure 5. Annual deposition trend of the total deposition.

5. CONCLUSION

The paper shows that for long periods of time the part of sulphur pollution, released in one and deposed over other countries and territories in Southeast Europe is significant. The obtained results are in good agreement with former calculations by other authors (Ganev and Syrakov, 2002). In the comparison with officially published results from the status report of EMEP/MSC-W that the exchange of sulphur pollution between these countries is estimated in the correct order of magnitude, giving at the same time much more details in the time and space distribution of deposed quantities. Generally, the emissions, respectively the concentrations and depositions, decreased during the period, although with relative small rate. The author’s conclusion is that the model produces a reasonable picture of the concentrations and depositions in the 25-km grid for the sulphur components in the region of Balkans.
The results of such calculations can be used in decision-making, negotiating and contamination strategies development.

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