Computer simulations of the atmospheric composition climate of Bulgaria

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Abstract
Some extensive numerical simulations of the atmospheric composition fields in Bulgaria have been recently performed. The US EPA Model-3 system was chosen as a modelling tool. As the NCEP Global Analysis Data with 1 degree resolution was used as meteorological background, the MM5 and CMAQ nesting capabilities were applied for downscaling the simulations to a 3 km resolution over Bulgaria.
The TNO emission inventory was used as emission input. Special pre-processing procedures are created for introducing temporal profiles and speciation of the emissions. The biogenic emissions of VOC are estimated by the model SMOKE. The simulations were carried out for years 2000-2007.
The numerical experiments have been carried out for different emission scenarios, which makes it possible the contribution of emissions from different source categories to be evaluated. The Models-3 "Integrated Process Rate Analysis" option is applied to discriminate the role of different dynamic and chemical processes for the air pollution formation.
The obtained ensemble of numerical simulation results is extensive enough to allow statistical treatment – calculating not only the mean concentrations and different source categories contribution mean fields, but also standard deviations, skewness, etc. with their dominant temporal modes (seasonal and/or diurnal variations). Thus some basic facts about the atmospheric composition climate of Bulgaria can be retrieved from the simulation ensemble.
Key words: air pollution modelling, US EPA models-3 system, multi-scale modelling, emission scenarios, process analysis

Simulaciones computacionales del clima de Bulgaria y su composición atmosférica

Resumen
Se han realizado simulaciones numéricas con el sistema US EPA Model-3 sobre todo el territorio de Bulgaria con una resolución horizontal de hasta 3 km en el periodo 2000-2007.
Los input necesarios para activar el sistema US EPA Model-3 son, básicamente dos, datos meteorológicos del NCEP (NCEP Global Analysis Data) con 1º de resolución, y el inventario de emisiones del TNO. Aunque las emisiones biogénicas de los compuestos orgánicos volátiles (COV's) se estimaron mediante modelo SMOKE. Para poder introducir las emisiones al modelo US EPA Model-3 se realizó un procesado previo que permitía determinar sus perfiles temporales y las especificaciones para cada una de las emisiones. En las simulaciones realizadas se consideraron diferentes escenarios de emisiones de diferentes tipos de fuente.

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Para ver la contribución de cada los procesos dinámicos y químicos en los niveles de contaminación se consideró la opción “Integrated Process Rate Analysis” del sistema US EPA Model-3.
Los resultados obtenidos de las simulaciones se analizaron estadísticamente y se determinaron las concentraciones medias, la contribución de las diferentes fuentes a los campos medios, la desviación estándar, el sesgo, …, así como los modos temporales dominantes (variaciones estacionales y/o diurnas).
Palabras clave: modelización de contaminación atmosférica, sistema US EPA models-3, modelización multiescala, escenarios de emisiones, análisis de los procesos.

Contents:

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1. Introduction

Recently extensive studies for long enough simulation periods and good resolution of the atmospheric composition status in Bulgaria have been carried out using up-to-date modeling tools and detailed and reliable input data (Gadzhev et al. 2011; 2012; 2013 a,b; 2014 a,b,c).

The simulations aimed at constructing of ensemble, comprehensive enough as to provide statistically reliable assessment of the atmospheric composition climate of Bulgaria – typical and extreme features of the special/temporal behavior, annual means and seasonal variations, etc.

The present paper, in which a brief review of the studies, will focus on some important characteristics of the atmospheric composition climate of Bulgaria – the concentrations of different compounds and the evaluation of the contribution of different emission categories to the overall air pollution in the country.

The air pollution pattern is formed as a result of interaction of different processes, so knowing the contribution of each for different meteorological conditions and given emission spatial configuration and temporal behavior is by all means important That is why the one of the important issues in the present paper is to present some evaluations of the contribution of different processes to the regional pollution over Bulgaria.

2. Brief description of the computer simulations

The simulations are based on the US EPA Model-3 system. The system consists of three components: MM5 (Dudhia 1993, 1996; Dudhia et al. 2005; Grell et al. 1994), used as meteorological pre-processor, CMAQ (Byun et al. 1998; Byun and Ching 1999), the Chemical Transport Model of the system and SMOKE (CEP 2003) – the emission pre-processor of Models-3 system.

The CMAQ “Integrated Process Rate Analysis” option was applied to discriminate the role of different dynamic and chemical processes for the air pollution pattern formation. The procedure allows the concentration change for each compound for an hour Δc to be presented as a sum of the contribution of the processes, which determine the concentration.

The large scale (background) meteorological data used by the study is the NCEP Global Analysis Data with 1º×1º resolution. The MM5 and CMAQ nesting capabilities are used to downscale the problem to a 3 km horizontal resolution for the innermost domain (Bulgaria). The simulations are carried out for fifteen sigma levels, the upper one being at approximately 16070m.

The TNO high resolution emission inventory (Visschedijk et al., 2007) is exploited. A detailed description of the emission modeling is given in Gadzhev et al. (2013a).

The study is based on a large number of numerical simulations carried out day by day for years 2000-2007 and five emission scenarios – with all the emissions and with biogenic emissions (categorie BB in Fig. 5 and 6), emissions from energetics (categorie SN_1), road transport (categorie SN_7), and none industrial combustion reduced (categorie SN_2). This makes it possible to evaluate the contribution of different emission categories to the formation of the overall atmospheric composition pattern. Performing extensive simulations of this kind with up to date highly sophisticated numerical models obviously requires large computer resource. That is why grid computing (Foster and Kesselmann, 1998; Atanasov. et al., 2006) was applied for the present simulations. Details about the performance of this grid application can be seen in Gadzhev et al. (2013a).

3. Validation of the computer simulation results

The computer simulations were validated by comparison with data of the pollution levels, measured by the Bulgarian National Network for Air Quality Control (Gadjhev 2014c).

Scatter diagrams of simulated and measured ozone levels for some arbitrarily taken stations are demonstrated in Gadjhev (2014c). They show that almost all the points are within the FA2 margins, which means that the condition for no more than 50% uncertainty of the hourly ozone values, defined in the respective European directive (European Parliament 2002) is fulfilled. The simulated results tend to underestimate the high ozone values and to overestimate the low ones.

The running 8-hour average values for simulated and measured ozone concentrations have been also calculated. The respective scatter diagrams (Gadjhev 2014c) show that the agreement between the simulated and measured running 8-hour average ozone values is much better in comparison to the hourly values with less dispersion around the ideal correspondence line and the better correlation is obvious. The above quoted requirement for less than 50% uncertainty is strictly fulfilled.

Some statistical evaluations of the O_3 and NO_2 simulations performance are shown in Tables 1, 2 for 14 monitoring stations, shown in Figure 1. The total number of stations, placed in 34 settlements all over the country - urban, living, high traffic, and industrial areas is 53. The stations, shown in Fig. 1 are chosen, because they only have ozone measurements and the data records for the simulated period are comprehensive enough.

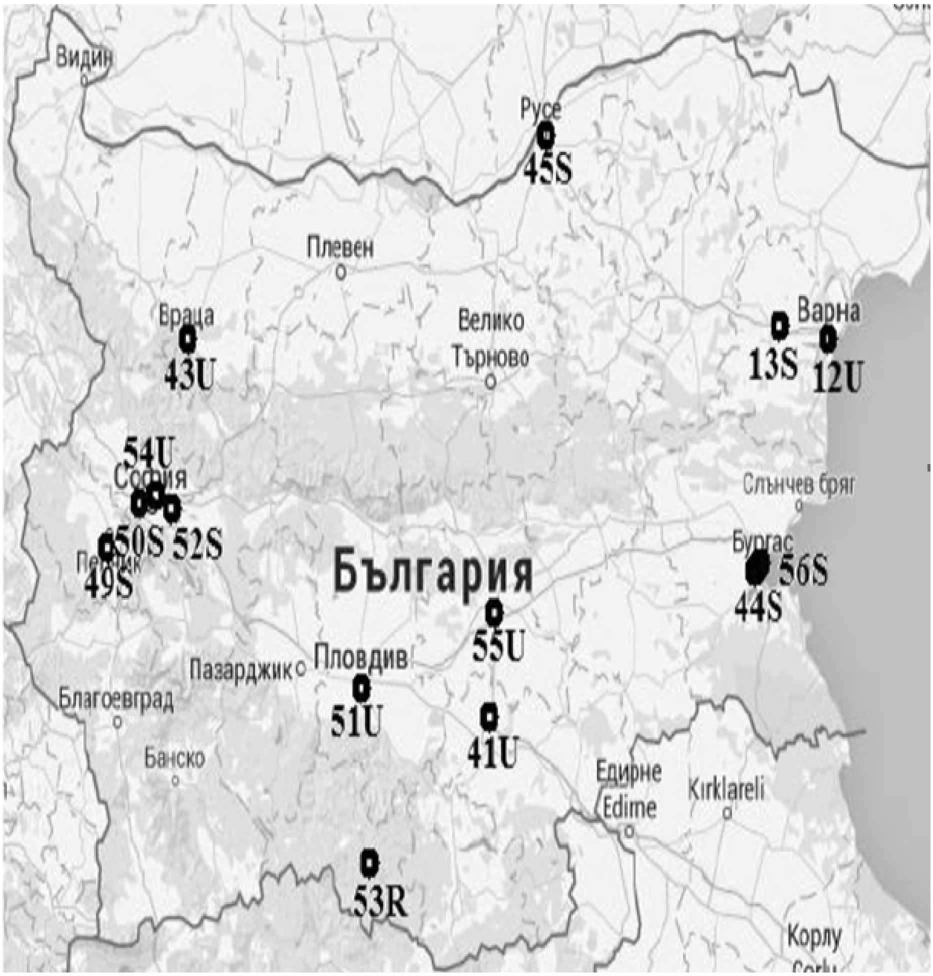

Figure 1. Map with the names, codes and emplacement of the used Bulgarian stations (U-urban, S-suburban, R-rural) 12U - Varna, 13S - Devnia, 41U - Dimitrovgrad, 43U - Vratza, 44S - Burgas, 45S - Ruse, 49S - Pernik, 50S - Krasno Selo -Sofia, 51U - Plovdiv, 52S - Drujba -Sofia, 53R - Rojen peak, 54U - Orlov most-Sofia, 55U - Stara Zagora, 56S - Burgas.

Table 1. Some statistical evaluations of the simulated ensemble with measured data for O_3: MP, MO – mean simulated and observed concentrations, NMB – normalised mean bios, NRMSE – normalised root mean square error, FA2 - % of cases within FA2 margins, PCC – correlation coefficient, NMSD – normalised mean square deviation

Station	MP $\mu g/m^3$	MO $\mu g/m^3$	NMB %	NRMSE %	FA2 %	PCC	NMSD %
12U	71.45	72.41	1.35	11.27	87.30	0.45	-41.04
13S	72.49	70.25	-2.56	12.71	91.45	0.49	-45.36
41U	72.92	71.67	-1.72	11.71	90.76	0.67	-32.95
43U	69.68	76.69	10.05	15.19	82.77	0.52	-44.87
44S	73.72	72.47	-1.70	12.46	88.26	0.72	-44.54
45S	70.48	71.99	2.14	12.34	88.97	0.67	-36.67
49S	67.43	73.00	8.27	6.92	85.27	0.53	-32.70
50S	60.08	75.18	25.13	12.77	75.90	0.69	-12.63
51U	67.19	72.37	7.71	10.35	86.06	0.68	-31.19
52S	61.34	66.92	9.09	9.85	86.14	0.68	-9.23
53R	88.96	82.64	-7.11	6.42	98.76	0.58	-33.87
54U	66.70	67.72	1.53	8.84	88.15	0.72	-19.60
55U	61.61	72.11	17.05	16.59	80.81	0.55	-27.91
56S	80.34	74.19	-7.65	14.02	94.91	0.62	-46.14

Table 2. Some statistical evaluations of the simulated ensemble with measured data for NO_2: MP, MO – mean simulated and observed concentrations, NMB – normalised mean bios, NRMSE – normalised root mean square error, FA2 - % of cases within FA2 margins, PCC – correlation coefficient, NMSD – normalised mean square deviation

Station	MP $\mu g/m^3$	MO $\mu g/m^3$	NMB %	NRMSE %	FA2 %	PCC	NMSD %
12U	15.47	7.11	-54.04	7.52	50.84	0.52	-57.49
13S	16.87	8.23	-51.22	5.07	53.82	0.38	-64.49
41U	25.32	11.10	-56.15	9.47	43.67	0.35	-68.54
43U	12.83	5.85	-54.45	9.63	49.29	0.51	-60.45
44S	9.98	5.99	-39.96	8.73	62.17	0.63	-38.77
45S	13.85	6.04	-56.35	9.25	49.21	0.46	-71.51
49S	22.66	9.51	-58.02	15.03	43.58	0.42	-56.48
50S	23.45	10.14	-56.76	9.56	43.20	0.47	-54.84
51U	18.55	7.48	-59.68	9.09	46.62	0.47	-74.09
52S	27.28	16.91	-38.01	7.57	64.52	0.67	-42.91
53R	3.83	2.71	-29.22	10.69	75.52	0.71	-39.38
54U	42.07	21.44	-49.04	10.07	52.10	0.65	-44.92
55U	14.01	5.12	-63.42	7.15	42.76	0.46	-78.92
56S	7.59	4.10	-45.91	7.84	56.96	0.61	-50.69

Criteria of acceptance of the simulated/measured concentrations agreement are defined in Thunis et al. (2013a, b). The comparison of the results in Tables 1, 2 with these criteria, shows that for most of the stations the criteria are fulfilled. The NO_2 simulations, in particular evaluated by the FA2 criterion, perform worse. This can be explained partially by the great uncertainty in the NO_2 emission inventory – the NO_2 emissions from road transport are given as total for the country and their spatial distribution is determined by surrogates – the road categories and network density.

The other probable reason is that the stations of the Bulgarian National Network for Air Quality Control are mostly located in the cities and near big industrial sources in order to reflect the highest pollution levels. The simulation horizontal spatial resolution (3 km) is probably not good enough to "catch" these NO_2 maxima. The ozone fields, from the other hand, are smoother, with smaller horizontal gradients and maxima not so closely related to the sources.

The comparison of the simulated fields with data of the pollution levels shows an agreement, which is not brilliant. The acceptance criteria, defined in Thunis et al. (2013a, b) are, however, fulfilled to a great extend. This means that the agreement is reasonable enough, so that the simulated ensemble can be treated as representative reliable for the atmospheric composition climate of Bulgaria.

4. Some examples, comments and discussion

The most simple atmospheric composition evaluations are, of course, the surface concentrations. By averaging over the 8-year simulated fields ensemble the mean annual and seasonal surface concentrations can be obtained and treated as respective "typical" daily concentration patterns.

Plots of some of these "typical" annual surface concentrations are shown in Fig. 2 for some of the most popular compounds – NO_2, SO_2, ozone. What can be seen from the plots is not surprising: the big cities and the road network are clearly outlined in the NO_2 surface concentrations, the big power plants in the SO_2 surface concentrations.

The ozone fields are much more complex. What should be mentioned is the expected effect of ozone minimums over big cities. The road network can also be followed in the plots as lines with lower ozone concentrations. This is in a good agreement with the ozone chemistry scheme.

The seasonal and diurnal variations of the averaged for the country surface O_3 and NO_2 are shown in Fig. 3, together with the mean, maximal and minimal values there are also the curves denoted by 0.25, 0.75, 0.1 and 0.9. These curves show the imaginary concentrations for which the probability of the simulated ones to be smaller is respectively 0.25, 0.75, 0.1 and 0.9. Thus the band 0.25-0.75 contains 50% and the band 0.1-0.9 - 80% of the possible cases. The plots are self explanatory enough and demonstrate the seasonal and diurnal O_3 and NO_2 variations.

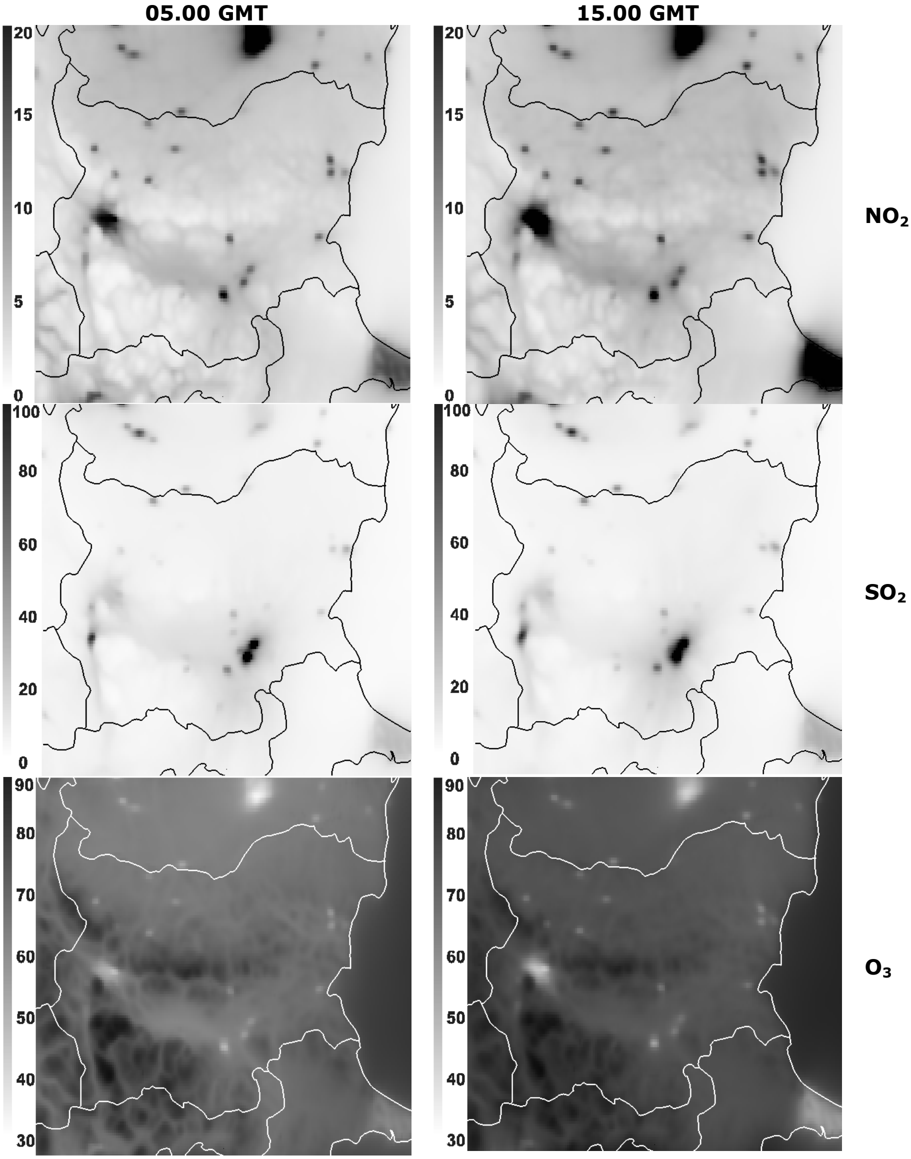

Figure 2. Surface concentrations of NO_2, SO_2 and O_3 [$\mu g/m^3$] averaged annually at 05.00 and 17.00 GMT

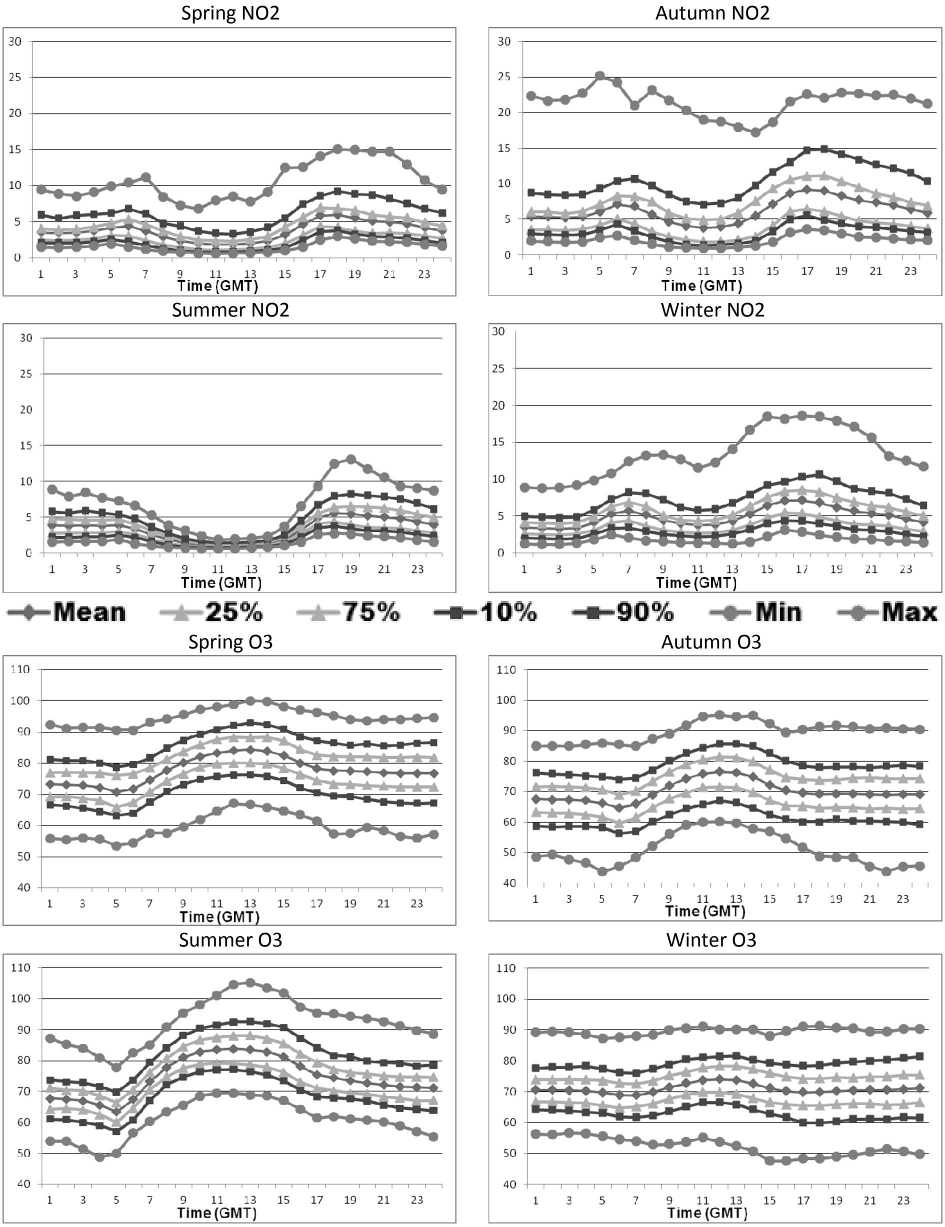

Figure 3 Seasonal variations of the O_3 and NO_2 surface concentrations [$\mu g/m^3$], averaged for the territory of Bulgaria: curves of mean, maximal and minimal values as well as curves show the imaginary concentrations for which the probability of the simulated ones to be smaller is respectively 0.25, 0.75, 0.1 and 0.9.

The local effects on the ensemble behaviour are demonstrated in Fig. 4, where the same characteristics are calculated for Bulgaria, Rojen (mountain site) and the cities of Sofia and Stara Zagora (smaller city). As it can be seen, the local effects are also very well displayed, in particular in the NO_2 fields, which for the different places simply can not be plotted with the same scale. The geographical variations in the O_3 behaviour are much smaller.

An important characteristic of the atmospheric composition climate of the country is the contribution of the emission of different categories to the overall atmospheric composition pattern. The relative contributions were calculated day by day and then, by averaging over the 8-year ensemble the "typical" contributions for the four seasons and annually were obtained. Some illustrations of the emission impact evaluations will be given in the present paper.

In order to demonstrate the emission contribution behaviour in a more simple and easy to comprehend way, the relative contribution fields can be averaged over some domain (in this case the territory of Bulgaria), which makes it possible to jointly follow and compare the diurnal behaviour of the respective contributions for different species. Such plots for some of the compounds are given in Fig. 5.

There is no need to describe the plots in details, but some comments on them could be made. First of all it could be seen that the different emissions relative contribution to the concentration of different species could be rather different. The contributions of different emission categories to different species surface concentrations have different diurnal course and different importance. The energy production is the major contributor to SO_2 and PM2.5 concentrations, while the biogenic emissions have near zero or even negative contributions. The major contributors to the NO_2 concentrations are the road transport and biogenic emissions. Their diurnal courses are in counter-phase, which can be easily explained by the ozone photochemistry cycle.

One can not help but notice the small contribution of biogenic emissions to surface ozone. This fact was extensively discussed in Gadzhev (2012, 2013a) and was explained by the fact that for Bulgaria the local O_3 production rate is limited by the availability of NO_x, a regime which is called NO_x - limited. The contribution of the emission from categories 1 and 7, which are the major sources of the other ozone precursor – nitrogen oxides, is also small. This is an indirect indicator, that the surface ozone in Bulgaria is to a small extend due to domestic sources, but is mostly imported.

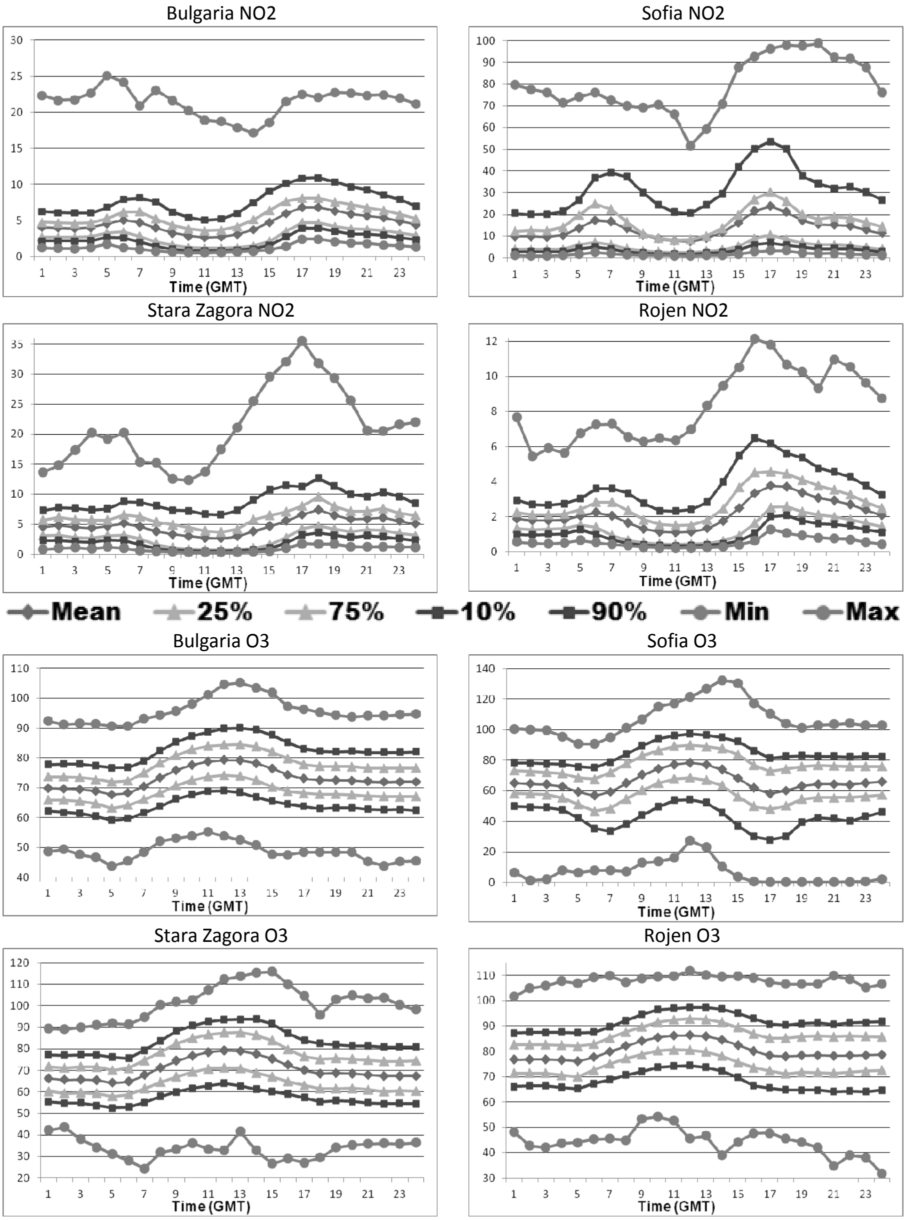

Figure 4 Geographical variations of the annual mean O_3 and NO_2 surface concentrations [$\mu g/m^3$] - averaged for the territory of Bulgaria and for Rojen, Sofia and Stara Zagora: curves of mean, maximal and minimal values as well as curves show the imaginary concentrations for which the probability of the simulated ones to be smaller is respectively 0.25, 0.75, 0.1 and 0.9.

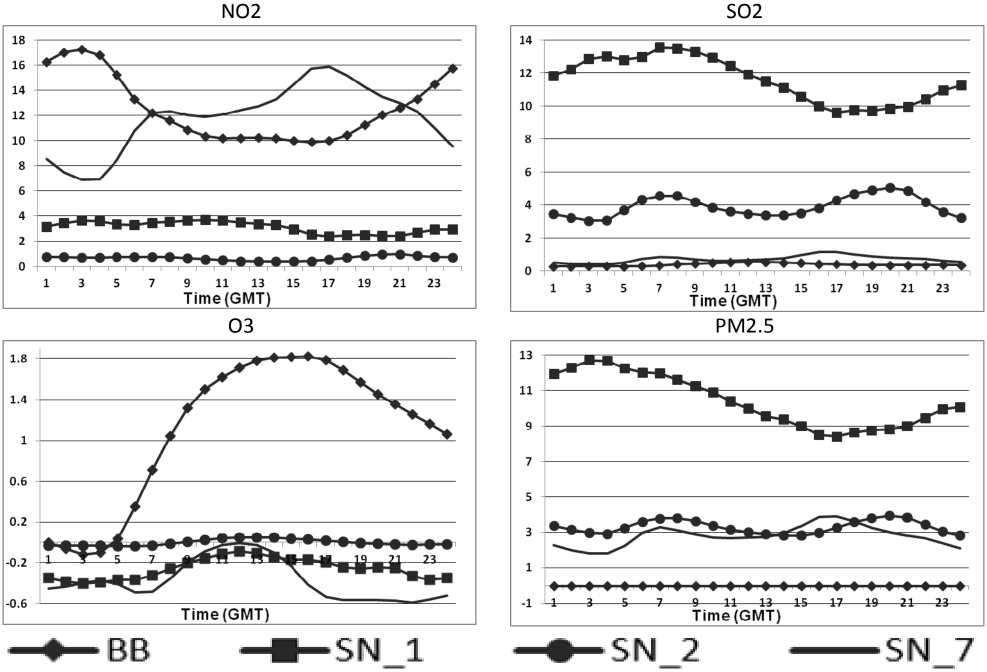

Figure 5 Plots of the "typical" annual diurnal course of the averaged for the territory of Bulgaria relative contributions [%] of emissions from categories 1 (SN_1), 2 (SN_2) and 7 (SN_7) and of the biogenic emissions (BB) to the concentrations of NO_2, SO_2, O_3 and PM2.5.

The picture is completely different for the city of Sofia (Fig.6). The NO_2 concentrations are totally dominated by road transport emissions. The none-industrial combustion has big contribution in SO_2 formation (probably mostly from the city heating plants and domestic heating). The NO_2 also has dominating (negative) contribution to the surface ozone. It is particularly large in morning and late afternoon, when the city traffic is most intensive. In the afternoon the contribution of road transport to the PM2.5 levels becomes even bigger than the contribution of the energy production emissions.

The different emission categories contributions for a typically mountain location (Rojen) are also given in Fig. 6. It can be seen that the behavior of the different emission categories id different compared to the urban location of Sofia. For example, the large contribution of biogenic emissions and the positive contribution of emissions from road transport around noon can be mentioned.

Another very important atmospheric composition characteristic is the contribution of different processes to the regional pollution over Bulgaria.

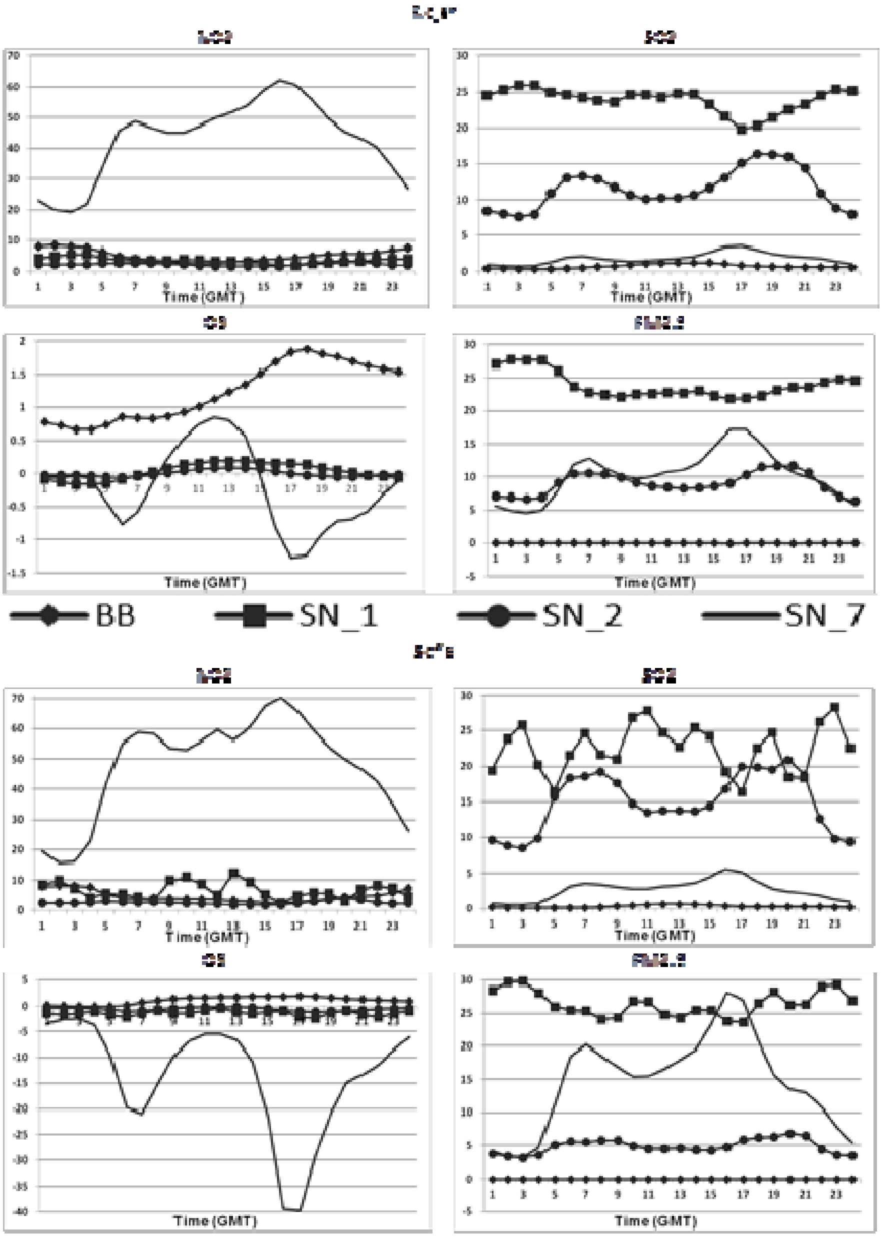

Figure 6. Plots of the "typical" annual diurnal course of relative contributions [%] of emissions from categories 1 (SN_1), 2 (SN_2) and 7 (SN_7) and of the biogenic emissions (BB) to the concentrations of NO_2, SO_2, O_3 and PM2.5 for Sofia and Rojen.

An example of the annually averaged special distribution of the processes contribution to the surface ozone is given in Fig. 7. It can be seen that the chemical processes have mostly negative impact. In particular the big cities and the road network (powerful nitrogen oxide sources) can be clearly followed as ozone sinks.

The vertical diffusion impact is mostly positive (turbulent transport of ozone from the upper layers). The effect is very prominent in the big cities, where the very large nitrogen oxide surface sources cause big ozone deficiency (big negative vertical gradients) and so the turbulent transport is more intensive. Some small spots of vertical diffusion negative impact can be seen at the location of big power plants. This is probably due to the fact that these are high sources of nitrogen oxide, which cause ozone deficiency aloft, so the ozone vertical gradients near surface are positive.

The horizontal and vertical advection contributions pattern is very complex and clearly reflects landscape induced local circulation systems. The horizontal and vertical advection contributions have mostly opposite signs, which is a direct and apparent consequence of the atmosphere continuity equation.

The horizontal diffusion, as it should, acts for compensating the ozone deficiency and so is generally in counter-phase with the chemical processes.

The averaged over the territory of Bulgaria contributions of some of the processes to the surface ozone concentrations will be also demonstrated (Fig. 8).

Very briefly the main characteristics, which can be seen from the plots, are the following:

(1) There are well manifested seasonal differences and diurnal variations;

(2) The ozone concentration change is formed as a rather small sum of processes with larger values and different signs;

(3) Averaged for the territory of Bulgaria the impacts of horizontal diffusion and cloud processes/aqueous chemistry are negligible;

(4) For all the seasons, except winter, and annually the vertical diffusion has a large positive impact, especially during the day (more intensive turbulence) – ozone transport from higher atmosphere to ground level;

(5) The dry deposition has negative impact, but it is almost negligible during winter and significant for spring, summer (in particular) and autumn during daytime. This is easy to explain – the dry deposition is proportional to surface concentration, and so is large when the surface concentrations are large;

(6) For all the seasons, except summer and especially in winter, and annually the horizontal advection has large positive impact. In summer around noon there is a period of horizontal advection negative impact. All this means that for most of the time there is ozone inflow trough the country boundary;

(7) The impact of chemical processes is always negative, except during daytime in the summer

The last three characteristic features of the processes behavior are sound evidence that the ozone/ozone precursors in Bulgaria are mostly of foreign origin.

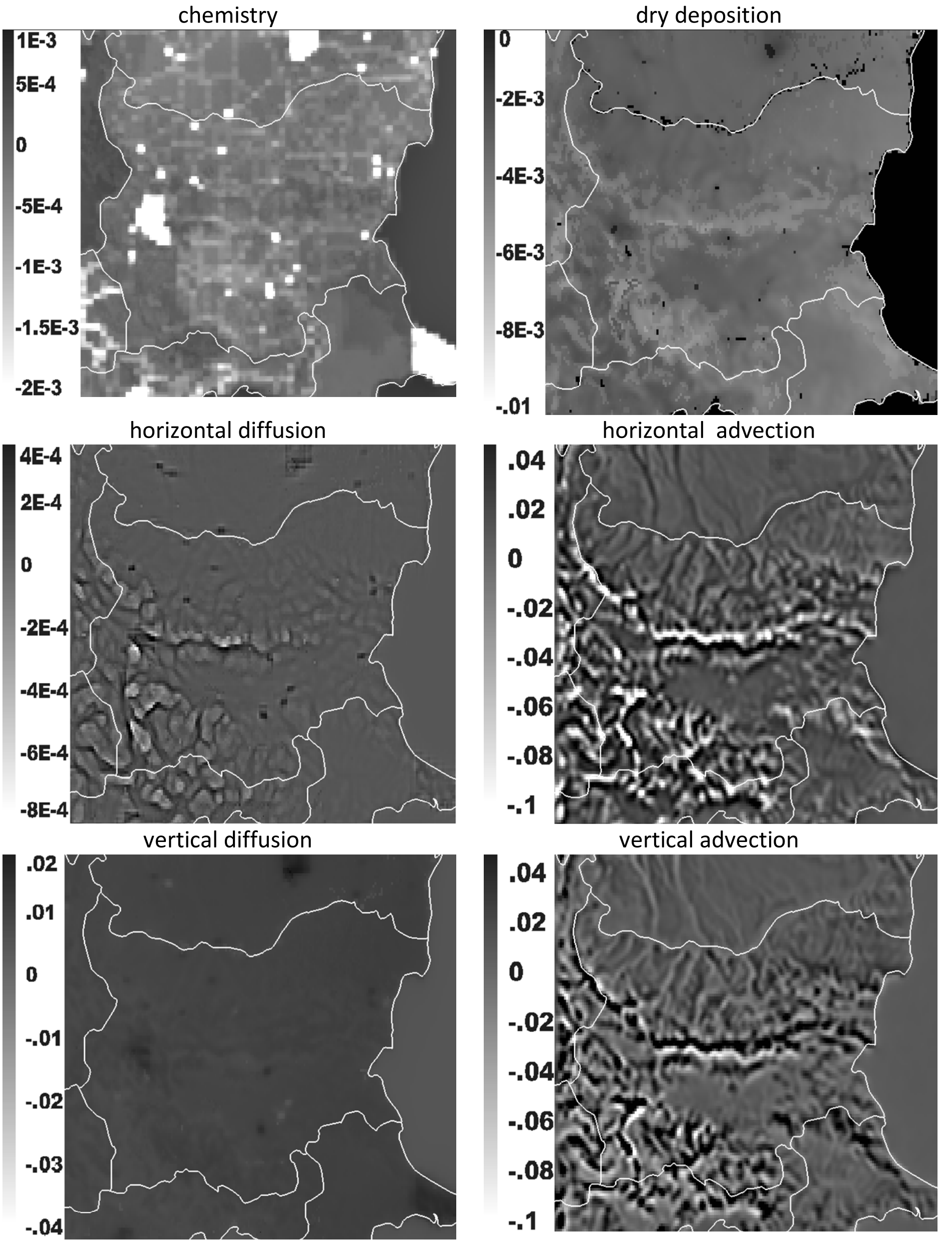

Figure 7 Horizontal distribution of the contributions [μg/hour] of different processes to the hourly surface ozone changes at 06.00GMT (08.00 local time).

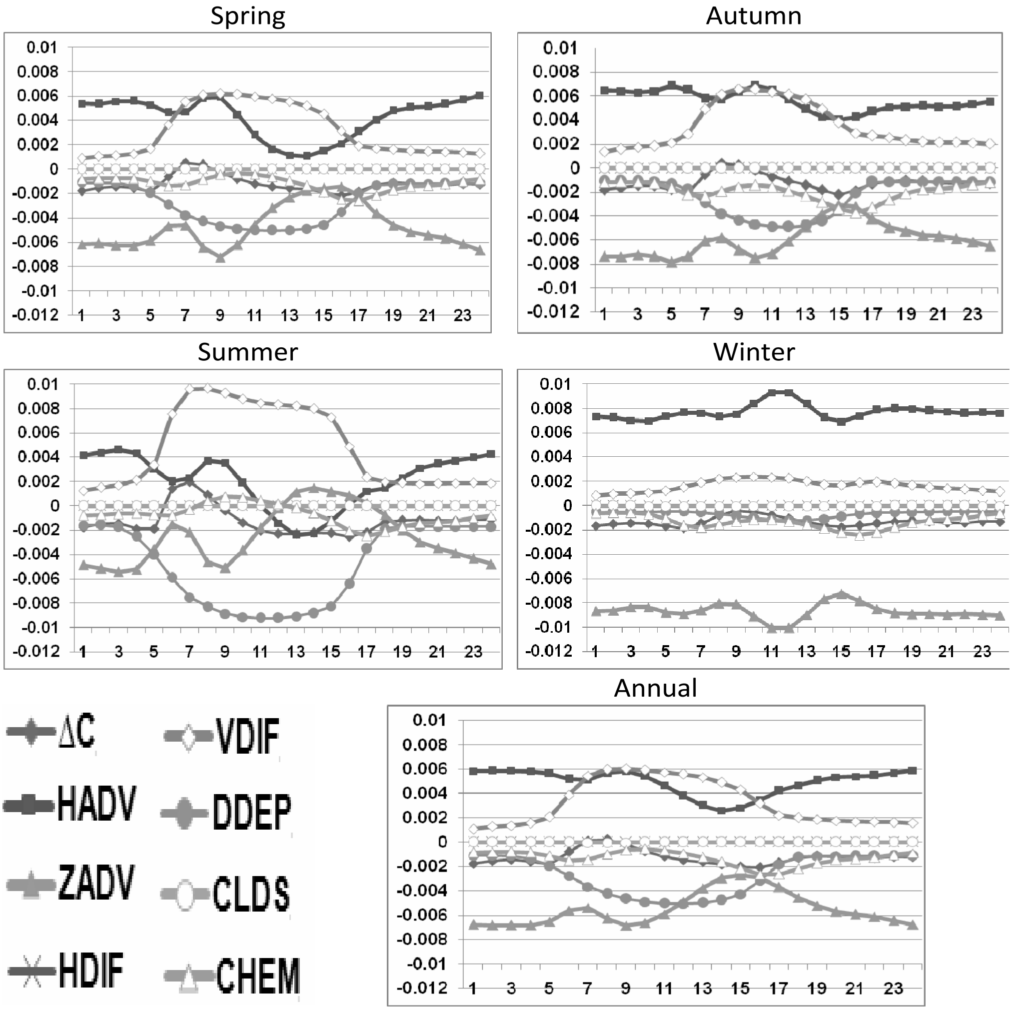

Figure 8 Plots of the "typical" annual and seasonal diurnal course of the averaged for the territory of Bulgaria contributions of vertical advection (ZADV), vertical diffusion (VDIF), horizontal diffusion (HDIF), dry deposition (DDEP), chemistry (CHEM), horizontal advection (HADV), cloud processes/aqueous chemistry (CLDS) to the hourly changes (Δc) of surface O_3.

In order the local heterogeneities of the different processes behavior to be demonstrated the annually averaged process contributions to SO_2 surface concentration changes for 4 different points in Bulgaria, together with the averaged for the country are shown in Fig. 9. It can be seen that the processes temporal behavior and interaction is different for the different points. It is remarkable how fast and chaotic the changes of the horizontal and vertical advection are for Sofia and Burgas. The horizontal and vertical advection contributions have mostly opposite signs, which effect had already been mentioned above. The diurnal course of horizontal and vertical advection contributions for Rojen is a very typical and good example of the role of mountain circulation.

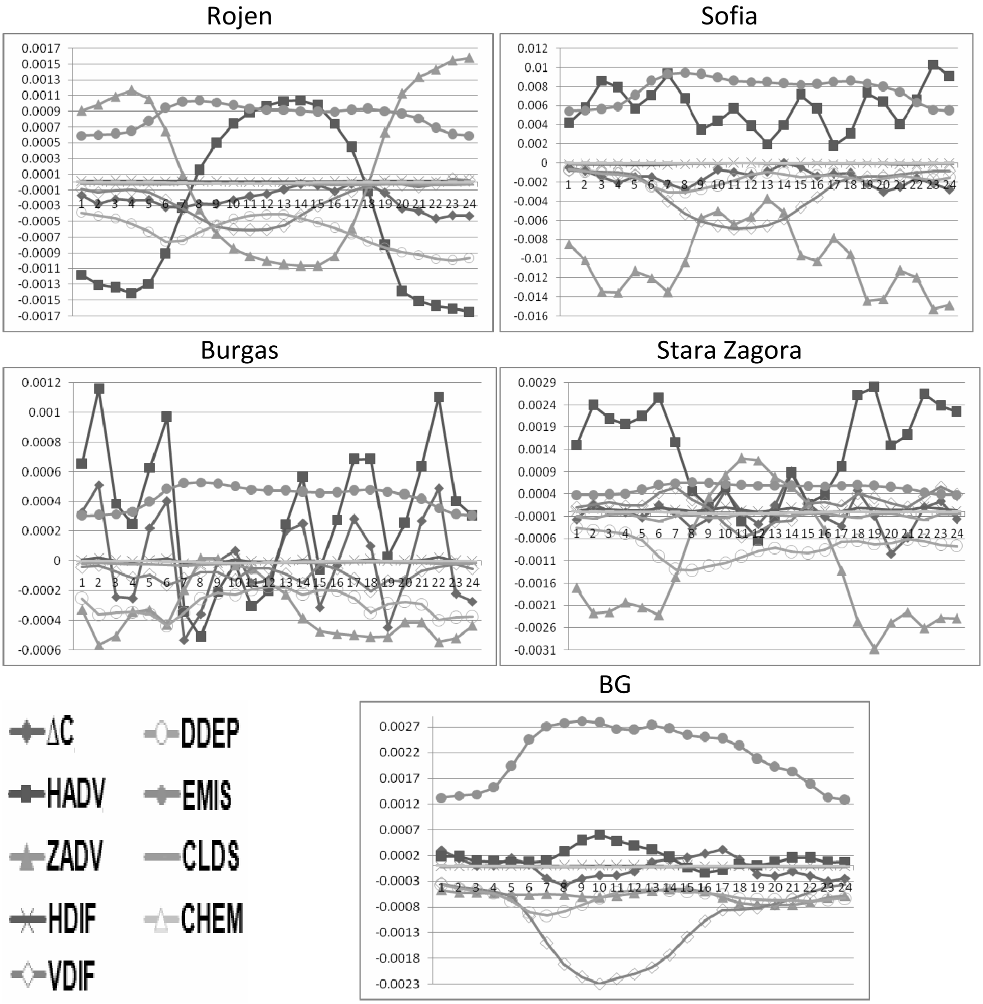

Figure 9. Plots of the "typical" annual diurnal course of the in some points and averaged for the territory of Bulgaria contributions of vertical advection (ZADV), vertical diffusion (VDIF), emissions (EMIS), dry deposition (DDEP), chemistry (CHEM), horizontal advection (HADV), horizontal diffusion (HDIF), cloud processes/aqueous chemistry (CLDS) to the hourly changes (Δc) of surface SO_2.

5. Conclusions

The numerical experiments performed produced a huge volume of information, which have to be carefully analyzed and generalized so that some final conclusions could be made. Nevertheless, some of the major findings so far will be listed below:

- the behavior of the surface concentrations, averaged over the ensemble annually, or for the four seasons and over the territory of the country is reasonable and demonstrates effects which for most of the compounds can be explained from a point of view of the generally accepted schemes of dynamic influences (in particular the role of turbulent transport and its dependence on atmospheric stability) and/or chemical transformations;
- the SN_1 contribution to the surface SO_2 concentrations is smaller than one should expect, having in mind that the “Maritza” power plants are among the biggest sulfur sources in Europe. Probably, a significant amount of SO_2 from these sources becomes a subject of larger scale transport and so is moved outside the country;
- the contribution of biogenic emissions to surface ozone in the country is relatively small. This indicates that local O_3 production rate is limited by the availability of NO_x concentration, a regime which is called NO_x-limited. Obviously from a point of view of atmospheric composition climate the Balkan Peninsula and Bulgaria are predominantly “rural” environment which explains the ozone photochemistry specifics in the region.;
- the contribution of the emission from categories 1 and 7, which are the major sources of the other ozone precursor – nitrogen oxides, is also small. This, once again is an indirect indicator, that the surface ozone in Bulgaria is to a small extend due to domestic sources, but is mostly imported;
- the results produced by the CMAQ “Integrated Process Rate Analysis” demonstrate the very complex behavior and interaction of the different processes. The analysis of the behavior of different processes does not give simple answer of the question how the air pollution in a given point or region is formed.

Acknowledgements

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References

ATANASSOV E., T. GUROV & A. KARAIVANOVA (2006): Computational Grid: structure and Applications, *Journal Avtomatica i Informatica* (in Bulgarian), year XL, September 2006, 40-43.

BYUN, D., J. YOUNG., G. GIPSON, J. GODOWITCH, F.S. BINKOWSKI, S. ROSELLE, B. BENJEY, J. PLEIM, J. CHING, J. NOVAK, C. COATS, T. ODMAN, A. HANNA, K. ALAPATY, R. MATHUR, J. MCHENRY, U. SHANKAR, S. FINE, A. XIU & C. JANG (1998) Description of the Models-3 Community Multiscale Air Quality (CMAQ) Modeling System*, 10th Joint Conference on the Applications of Air Pollution Meteorology with the A&WMA, 11-16 January 1998, Phoenix, Arizona*, 264-268.

BYUN D. & J. CHING (1999) Science Algorithms of the EPA Models-3 Community Multiscale Air Quality (CMAQ) Modeling System. *EPA Report 600/R-99/030, Washington DC*. http://www.epa.gov/asmdnerl/models3/doc/science/science.html.

CEP (2003) Sparse Matrix Operator Kernel Emission (SMOKE) Modeling System, University of Carolina, Carolina Environmental Programs, Research Triangle Park, North Carolina.

DUDHIA J. (1993) A non-hydrostatic version of the Penn State/NCAR Mesoscale Model: validation tests and simulation of an Atlantic cyclone and cold front. *Mon. Wea. Rev.* 121, 1493-1513.

DUDHIA J. (1996) A multi-layer soil temperature model for MM5, *Preprint from the Sixth PSU/NCAR Mesoscale Model Users' Workshop*

DUDHIA J., D. GILL, K. MANNING, WEI WANG, & G. BRUYERE (2005) *PSU/NCAR mesoscale Modeling System, Tutorial Class Notes and User's Guide: MM5 Modeling System Version 3*, http://www.mmm.ucar.edu/mm5/documents/MM5_tut_Web_notes/tutorialTOC.htm

European Parliament (2002) DIRECTIVE 2002/3/EC of 12 February 2002 relating to ozone in ambient air, *Official Journal of the European Communities* (9.3.2002), L67, 14-30.

FOSTER J. & C. KESSELMANN (1998) *The Grid: Blueprint for a New Computing Infrastructure*, Morgan Kaufmann.

GADZHEV G., G. JORDANOV, K. GANEV, M. PRODANOVA, D. SYRAKOV & N. MILOSHEV (2011) Atmospheric Composition Studies for the Balkan Region, *Lecture Notes in Computer Sciences, Dimov, I. S. Dimova, and N. Kolkovska (Eds.): LNCS 6046*, Springer-Verlag Berlin Heidelberg, 150-157.

GADZHEV G., K. GANEV, D., SYRAKOV, N., MILOSHEV & M., PRODANOVA (2012) Contribution of Biogenic Emissions to the Atmospheric Composition of the Balkan Region and Bulgaria, *Int. J. Environment and Pollution*, 50, 1/2/3/4, 130-139.

GADZHEV G., K. GANEV, N., MILOSHEV, D., SYRAKOV & M., PRODANOVA (2013a) Numerical Study of the Atmospheric Composition in Bulgaria, *Computers and Mathematics with Applications* 65, 402-422.

GADZHEV G., K. GANEV, D., SYRAKOV, M. PRODANOVA & N., MILOSHEV (2013b) Some Statistical Evaluations of Numerically Obtained Atmospheric Composition Fields in Bulgaria, Proc*eedings of 15th International Conference on Harmonisation within Atmospheric. Dispersion Modelling for Regulatory Purposes. 6-9 May 2013, Madrid, Spain*, 373-377.

GADZHEV G., K. GANEV, N. MILOSHEV, D. SYRAKOV, & M. PRODANOVA (2014a) Some Basic Facts About the Atmospheric Composition in Bulgaria – Grid Computing Simulations, in I. *Lirkov et al. (Eds.): LSSC 2013, LNCS 8353*, 484–490

GADZHEV G., K. GANEV, N. MILOSHEV, D. SYRAKOV, & M. PRODANOVA (2014b) Analysis of the Processes Which Form the Air Pollution Pattern over Bulgaria, in *I. Lirkov et al. (Eds.): LSSC 2013, LNCS 8353*, 390–396

GADZHEV G., K. GANEV & N. MILOSHEV (2014c) Numerical study of the atmospheric composition climate of Bulgaria – validation of the computer simulation results, *Proc. of the 16th International Conference on Harmonisation within Atmospheric Dispersion Modelling for Regulatory Purposes 8-11 September 2014, Varna, Bulgaria*, 41-45,

GRELL, G.A., J. DUDHIA & D.R. STAUFFER (1994) A description of the Fifth Generation Penn State/NCAR Mesoscale Model (MM5). *NCAR Technical Note, NCAR TN-398-STR*, pp. 138

THUNIS P., D. PERNIGOTTI & M. GERBOLES (2013a) Model quality objectives based on measurement uncertainty. Part I: Ozone. *Atmospheric Environment* 79, 861-868

THUNIS P., D. PERNIGOTTI & M. GERBOLES (2013b) Model quality objectives based on measurement uncertainty. Part II: NO2 and PM10. *Atmospheric Environment* 79, 869-878

VISSCHEDIJK A. J. H., P.Y.J. ZANDVELD & H.A.C. DENIER VAN DER GON (2007) A High Resolution Gridded European Emission Database for the EU Integrate Project GEMS, *TNO-report 2007-A-R0233/B*, Apeldoorn, The Netherlands.