NUMERICAL STUDY OF THE ATMOSPHERIC COMPOSITION CLIMATE OF BULGARIA – VALIDATION OF THE COMPUTER SIMULATION RESULTS

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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 modelling tools and detailed and reliable input data (Gadzhev et al. 2011, 2012, 2013 a,b,c,d).
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 behaviour, annual means and seasonal variations, etc.
The numerical experiments performed produced a huge volume of information, which was used as a basis for evaluation and clarification of the atmospheric composition climate of Bulgaria. It is natural, in such a case, that the model results should be validated by comparison with measured data. The outcome of these comparisons is demonstrated and commented in the present paper

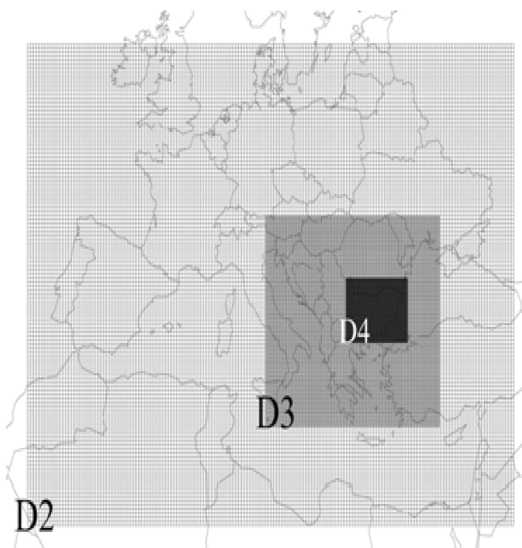
METHODOLOGY

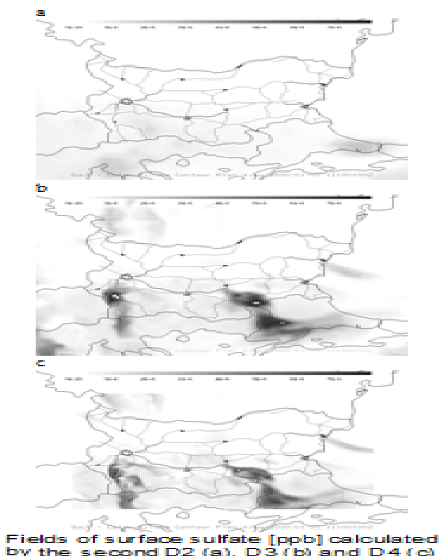
Domains and nesting

Basic models: US EPA Models-3 System

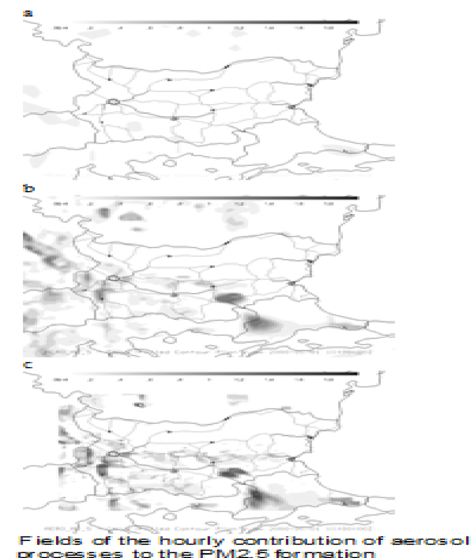
MM5 - the 5th generation PSU/NCAR Meso-meteorological Model MM5 used as meteorological pre-processor. This model is pretty often replaced by the next generation model WRF;
SMOKE - the Sparse Matrix Operator Kernel Emissions Modelling System – the emission pre-processor;
CMAQ - the Community Multiscale Air Quality System being the Chemical Transport Model (CTM);
The Models-3 "***Integrated Process Rate Analysis***" option is applied to discriminate the role of different dynamic and chemical processes for the air pollution pattern formation.
Data: The large scale (background) meteorological data used by the study is the NCEP Global Analysis Data with 1°×1° 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 TNO high resolution emission inventory is exploited. A detailed description of the emission modeling is given in Gadzhev et al. (2013a).
The MM5/CMAQ simulations were performed day by day for 8 years - from 2000 to 2007. Thus a quite extensive data base was created, which could be used for different studies and considerations of the main features and origins of the atmospheric pollution in Bulgaria.

	D1	D2	D3	D4
MM5 (km) 2 way nesting	81	27	9	3
CMAQ (km)		27	9	3
Grid dimensions		166X115	178x151	190x140





Fields of surface sulfate [ppb] calculated by the second D2 (a), D3 (b) and D4 (c) nesting steps, 01.01.2000, 11:00 UTC



Fields of the hourly contribution of aerosol processes to the PM2.5 formation [μg/(m³.hour)] calculated by the second D2 (a), D3 (b) and D4 (c) nesting steps, 01.01.2000, 11:00 UTC

RESULTS

The computer simulations were validated by comparison with data of the pollution levels, measured by the Bulgarian National Network for Air Quality Control.
Scatter diagrams of simulated and measured ozone levels for some arbitrarily taken stations in Figure 1. It can be seen, 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 are shown in Figure 2. It can be immediately seen that the agreement between the simulated and measured running 8-hour average ozone values is much better in comparison to the hourly values. The 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.

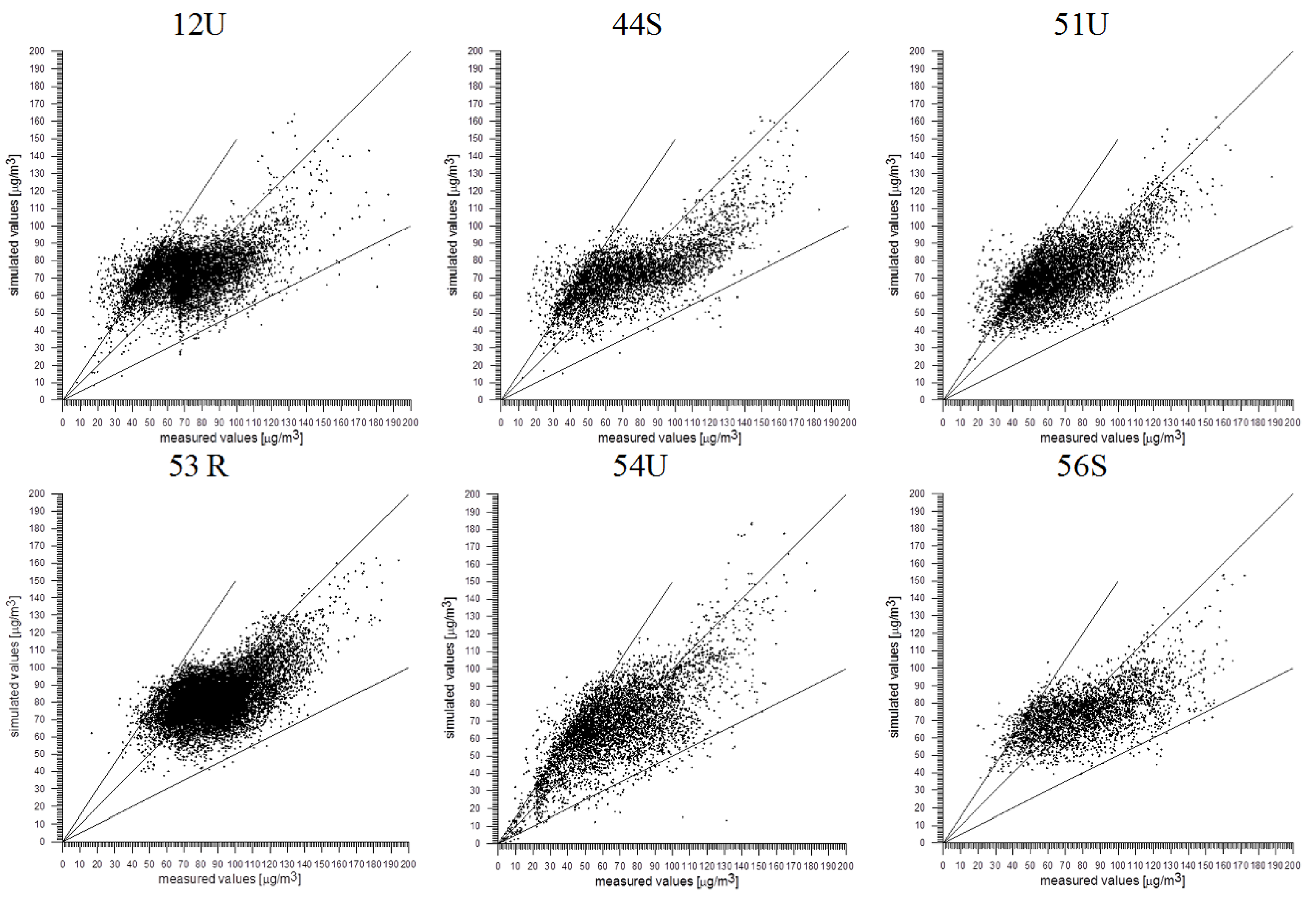


Figure 1. Scatter diagrams of simulated and measured ozone levels for some of the stations of the Bulgarian National Network for Air Quality Control.

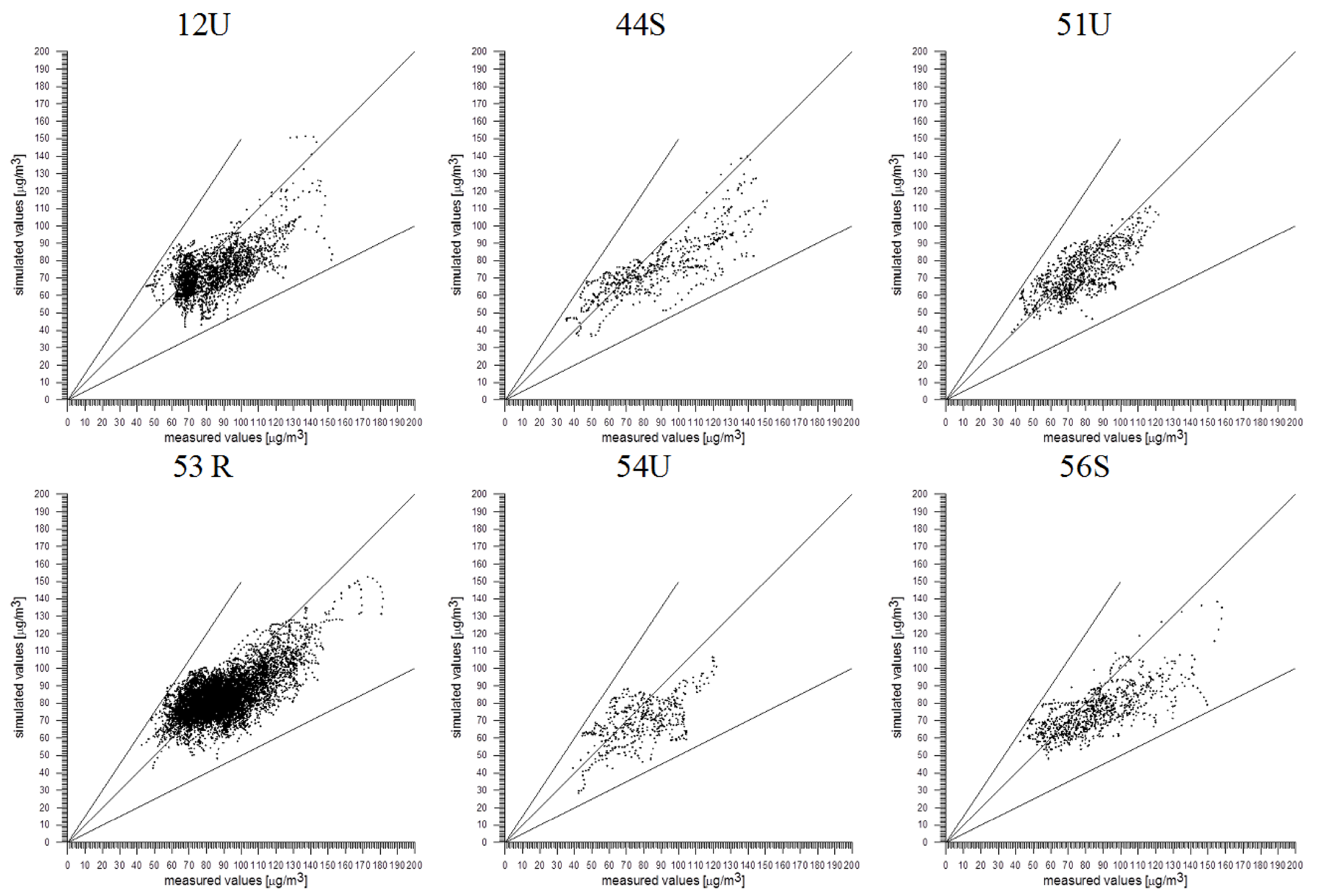


Figure 2. Scatter diagrams of running 8-hour average values for simulated and measured ozone levels for some of the stations of the Bulgarian National Network for Air Quality Control.

Some criteria of acceptance of the simulated/measured concentrations agreement are defined in Thunis et al. (2013z, 2013b). The comparison of the results in Tables 1, 2 with these criteria (Table 3), 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.

station	MO (μg/m³)	MP (μg/m³)	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 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	MO (μg/m³)	MP (μg/m³)	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

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

	O_3		NO_2	
	Rural	Urban / SubUrban	Rural	Urban / SubUrban
NMB	< 37%	< 41%	< 159%	< 79%
PCC	> 0.40	> 0.51	> 0.00	> 0.29
NMSD	< 107%	< 97%	< 200%	< 117%
FA2	> 50 %	> 75% / 77%	> 50 %	> 49.3% / 58.2%

Table 3. Acceptance criteria for O_3 and NO_2 simulation results

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CONCLUSIONS

The comparison of the simulated fields with data of the pollution levels, measured by the Bulgarian National Network for Air Quality Control shows an agreement, which is not brilliant. The acceptance criteria, defined in Thunis et al. (2013z, 2013b) 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. Thus the evaluations made in Gadzhev et al. (2011, 2012, 2013 a,b,c,d) about typical and extreme features of the special/temporal behaviour, annual means and seasonal variations of different pollution characteristics – concentrations, contribution of different source categories, contribution of different processes, etc. should be considered as valid enough to provide scientifically robust assessments of the atmospheric composition and its origin.
The comparison results are not thoroughly satisfying. As mentioned above, one of the certain reasons for the simulation errors is the uncertainty in the emission inventories. Solving this problem requires, however, not only research, but administrative efforts as well.

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