

ESTIMATION OF OZONE POLLUTION LEVELS IN SOUTHEAST EUROPE USING US EPA MODELS-3 SYSTEM

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ABSTRACT

Air quality modeling becomes an important tool for investigation of air pollution levels in different scales. In the paper, experiments are made as to determine the degree of reaction of the US EPA Models-3 system on change in emissions. Three domains are set over Europe with resolution 90-30-10 km, the last one embracing entirely Romania, Bulgaria and Greece. MM5 is run over these domains for June 2000 with NCEP 1x1 degree Global Analysis data. CMAQ is run on the two inner domains only. EMEP inventory grid data with 50 km resolution is used to prepare the emission input for the 30-km domain and its 16.67 km disaggregation - for the finer one. Proper time profiles, monthly, weekly and daily, are imposed upon yearly values. The CMAQ "default profiles" are used as initial and boundary conditions on the 30-km grid. A set of runs are performed with aqueous CB-IV chemical mechanism on. It includes a full emission base case run and three control emission scenarios of reduced NO_x(50%), VOC(50%) and combination NO_x(50%)-VOC(25%) as to check the influence of different geographical regions on ozone formation. Comparison between different runs and between the basic case calculations and measurements are shown and commented.

Key Words: Air Pollution Modelling, CMAQ Model, Ozone Pollution

1. INTRODUCTION

During the last two-three decades, the international collaboration decreased the threat of acid rains in almost all Europe. In place of acid rains new threats for the population, nature and historical heritage emerge. The increase of NO_x emissions and the photochemical processes in the atmosphere led to formation of high ozone levels at the ground surface. This is a relatively new problem, more severe in Western European countries, but promising to arise to threat in the East as well. The European ozone directives (EC, 1998, 1999, EP, 2002) are aimed to put limits to high level ozone exposure and to set common control strategy by assessing current and future air quality regulations designed to protect human health and welfare.

Air quality models provide powerful and reliable tools for performing such assessment. A big number of models and model systems with different level of complexity were developed, lately. Many of them can be found in the EEA Model Documentation System (<http://air-climate.eionet.eu.int/databases/MDS/index.html>) or in the respective site of US EPA (<http://www.epa.gov/scram001/tt22.htm#rec>).

Many scientists, especially those from Central and Eastern European countries, face a number of problems in dealing with air pollution matter – from environmental impact assessment to forecast of photochemical smog and formation of high ozone levels, when working to meet the requirements of respective EU Directives.

The forthcoming accession of Bulgaria to EU sets a number of problems related to the necessity of operating a contemporary air pollution modeling system. The choice, studying and implementation of such a tool to different regional and local problems were the aim of the 5thFP funded project BULAIR (<http://www.meteo.bg/bulair>). The first task of the team was to make an extensive review of the existing models and to choose suitable one/ones. It occurs that the Model-3 system of US EPA is one of the best modeling tools. It continues to be developed intensively by the efforts of a big community of scientists both in the US and Europe.

The system consists of three parts: the 5th generation PSU/NCAR Mesometeorological Model MM5 (Dudhia, 1993, Grell et al., 1994) used as meteorological pre-processor; the Sparse Matrix Operator Kernel Emissions (SMOKE) Modeling System (CEP, 2003) and the Community Multiscale Air Quality System CMAQ (Byun et al., 1998, Byun and Ching, 1999). Important advantages of this software are that it is free downloadable and can be run on contemporary PCs. In the same time, this is a modeling tool of large flexibility, with a range of options and possibilities to be used for different applications/purposes. Many research groups in Europe already use the Model-3 system or some of its elements and this number is increasing rapidly. In the light of the above, the main goal of this paper is to show the first results of implementing the MM5-CMAQ system for estimation of pollution levels in southeast Europe.

In Bulgarian National Institute of Meteorology and Hydrology, the PSU/NCAR mesoscale model **MM5**, non-hydrostatic version 3-6-1, and the EPA Models-3 CMAQ model, version 4.3, are installed on PC Dell dual CPU Xeon 2.8 GHz computer under RedHat Linux 7.3 operational system using Portland Group 4.02 FORTRAN compiler. In-depth descriptions of MM5 can be found in Dudhia (1993) and Grell et al. (1994), and at <http://box.mmm.ucar.edu/mm5/>. On the sight <http://www.epa.gov/asmdnerl/CMAQ/CMAQscienceDoc.html> all documentation of CMAQ is available. Free download of all Models-3 elements is available at <http://www.cmascenter.org/>.

2. MODELING DOMAIN

The MM5 preprocessing program TERRAIN is used to define three domains with 90, 30 and 10 km horizontal resolution (51×45, 79×61 and 160×103 grid points, respectively). These three nested domains are chosen in such a way that the finer resolution domain contains Bulgaria and two neighboring Balkan countries - Romania and Greece. Lambert conformal conic projection, with true latitudes at 30°N and 60°N, and the central point with coordinates 41.5°N and 24°E are chosen.

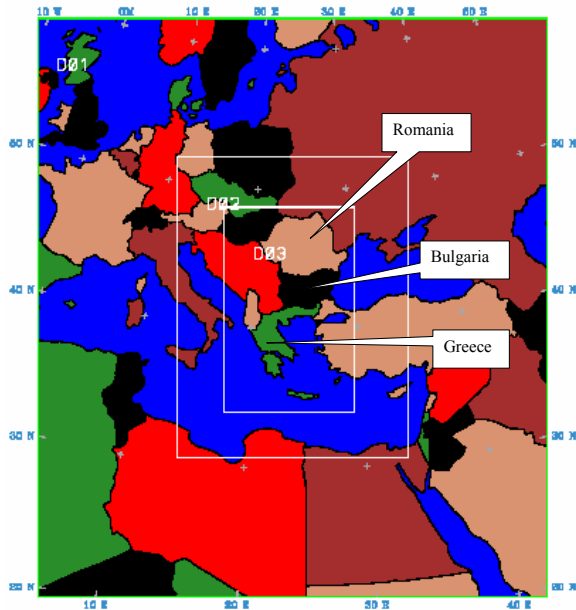


Fig. 1. The three computational domains

Together with grids definition TERRAIN specifies the raw topographic, vegetative, and soil type data to all grid points. The Land-Surface Model is included, so besides of elevation height and land-use data some additional fields such as enhanced soil types, vegetation fraction, and annual deep soil temperature are also generated. The model domains are shown in Fig. 1. The other pictures refer the inner grid only.

3. Meteorology

Meteorological database suitable for air quality modeling with CMAQ is generated by exploiting the US NCEP Global Analyses data for the year 2000 (<http://dss.ucar.edu/datasets/ds083.2/>). The data has 1×1 degree grid resolution covering the entire globe, the time resolution is 6 hours. NCEP derives these data from the FNL (final weather forecast run) which assimilates observations collected through at least six hours after the synoptic term. This FNL dataset has surface level, tropospheric levels, tropopause, and lower stratospheric levels analyses.

4. MM5 SIMULATION

CMAQ modeling guidance suggests that each grid on which the air quality model is to be run should be provided with MM5 meteorological fields that are developed independently in “one way” nesting mode. Here, CMAQ is run on the two finer grids only. So, MM5 is run on both outer grids (90 and 30 km) simultaneously with “two-way” nesting mode on, first. Then, after extracting the initial and boundary conditions from the resulting fields for the 10 km simulation, MM5 is run on the finer 10 km grid as a completely separate simulation with “one-way” nesting mode on. In this approach, information from the 30 km grid is transferred to the 10-km domain through boundary conditions during the simulation, but there is no feedback from the 10-km fields up-scale to the 30 km domain. All simulations are made with 23 σ -levels going up to 100 hPa height. Proper physical options are set in MM5. The model may be set to relax toward observed temperature, wind and humidity through four dimensional data assimilation, known as FDDA (Stauffer and Seaman, 1990). FDDA amounts to adding an additional term to the prognostic equations that serves to “nudge” the model solution toward the individual observations. This significantly reduces the drift in the solution for simulations of several days or more. The NCEP data set does not include observations, but analyzed data every 6 hours in all its grid points. MM5 is configured with FDDA option on as to nudge the model toward analyzed data on the 90 km grid only.

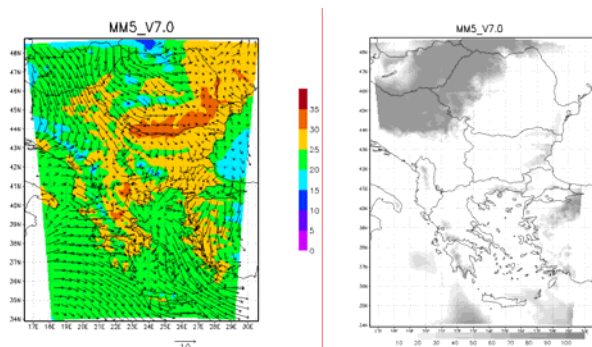


Fig. 2. Temperature (colored), wind and low cloud coverage on June 7, 2000, 14:00.

The 2000 annual simulation with MM5 is made on portions of 4 days. Every portion has additional 12 hours that are an initial spin-up period that overlaps the last 12 hours of the preceding run. Further on, the MM5 output for June is used to run CMAQ. In Fig.2, a temperature and wind combined map and a low level cloud cover map are shown.

5. EMISSION INPUT TO CMAQ

CMAQ demands its emission input in specific format reflecting the time evolution of all pollutants accounted for in the used chemical mechanism. The emission inventory usually is made on annual basis and many pollutants are estimated as groups (NO_x , SO_x , VOC - Volatile Organic Compounds). In preparing CMAQ emission file a number of specific estimates must be done. First, organic gases emissions estimates, and to a lesser extent SO_x and NO_x estimates, must be split, or “speciated”, into more defined compounds in order to be properly modeled for chemical transformations and deposition. Secondly, time variation profiles must be over-posed on these annual values to account for seasonal, weekly and daily variations. Finally, all this information must be gridded. The different types of sources (area, elevated point, mobile and biogenic) are treated in specific way. Obviously, emission models are necessary as reliable emission input to the chemical transport models to be produced. Such a component in EPA Models-3 system is SMOKE. Unfortunately, it is highly adapted to the US conditions - emission inventory, administrative division, motor fleet etc. Many European scientific groups are working now for adapting SMOKE to European conditions (see Borge et al., 2004).

Here, the CMAQ emission input is prepared exploiting the EMEP 50×50 km gridded inventory (Vestreng, 2001) for the entire EMEP area and its desagregation described in Ambelas Skjøth et al. (2002). The values over the grid points of the 30-km domain of this study are obtained by bi-linear interpolation over the 50 km EMEP gridded emissions and for the inner 10-km grid – over the desagregated 16.67-km inventory. Additional corrections are included for congruence between both inventories.

As to prepare the CMAQ emission input file the computer code **E_CMAQ** is created. The gridded (30 and 10 km) annual emission rates of 5 generalized pollutants – SO_x , NO_x , VOC, NH_3 and CO – are introduced in E_CMAQ and are speciated following the way recommended Smylie et al. (1991). The final speciation to 13 compounds, required by CB-IV chemical mechanism of CMAQ, is presented in Table 1:

Table 1. Speciation of “lumped” species as required by CB-IV.

No.	POLLUTANT	Split factor	Molecular Weight
	SO_x (as SO₂):		
1	SO ₂	0.85	64.065
2	SULF	0.15	96.
3	NH ₃		17.031
4	CO		28.01
	NO_x (as NO₂):		
5	NO ₂	0.15	46.006
6	NO	0.85	30
	VOC:		
7	OLE	0.087306	27.74
8	PAR	0.56023	13.87
9	TOL	0.139938	97.09
10	XYL	0.09172	110.96
11	FORM	0.01242	13.87
12	ALD2	0.018304	27.74
13	ETH	0.105674	27.74

The molecular weight of every compound is necessary for transforming individual emission rates from 1000tone/year to [mole/s] as required by CMAQ. The next modification of the so prepared data is the over-posing of proper time variation profiles (monthly, weekly and hourly). The methodology developed in USA EPA Technology Transfer Network (<http://www.epa.gov/ttn/chief/emch/temporal/index.html>) is adopted. As far as in the used gridded inventory the type of sources is not specified, some common enough area sources are chosen from the EPA SCC (Source Category Code) classification and their profiles are averaged. The resulting profiles are implemented. Finally, all the data, day by day, is written in the specific format required by CMAQ.

It must be stressed here that the biogenic emissions of VOC are not included in this study and this fact have to be taken into consideration when interpreting model results.

6. CMAQ SIMULATION

The CMAQ model requires inputs of three-dimensional gridded wind, temperature, humidity, cloud/precipitation, and boundary layer parameters. A meteorological database suitable for air quality modeling with CMAQ was generated by performing a year-long run with MM5 as described above. From this MM5 output CMAQ meteorological input was created exploiting the newest version of the CMAQ meteorology-chemistry interface - MCIP, v2.3. In the same time, the computational domains decrease by 6.5 points (boundary points) from every side and the dot- and the cross points exchange their place (that is why the half point appear).

CMAQ has been run from 00:00 UTC of 01 June to 00:00 UTC of 09 June, 2000, day by day on both domains at 6 vertical layers. The period is chosen without any specific consideration. The Chemical Transport Model has been run on both processors using an open-source, portable implementation of the Message-Passing Interface Standard (MPICH), version 1.2.5. The CB-4 chemical mechanism with Aqueous-Phase Chemistry and MEBI solver has been exploited.

The CMAQ pre-defined (default) concentration profiles are used for initial conditions over both domains at the beginning of the simulation. The concentration fields obtained at the end of a day's run are used as initial condition for the next day. Default profiles are used as boundary conditions of the 30-km domain during all period. The boundary conditions for the inner domain are determined through the nesting capabilities of CMAQ. Further on, results for the last 4 days of this period (5-8 June 2000) will be presented as to avoid the influence of the artificial initial conditions. This is the reason to interpret only the results on the inner 10-km domain.

7. SENSITIVITY OF CMAQ PHOTO-CHEMICAL CALCULATIONS

The elaboration of effective control strategy for decreasing ozone levels requires a knowledge of the response of ozone concentrations to the variation of its main precursors –NO_x and VOC. This response depends nonlinearly on spatially and temporarily variable factors. A big number of researches with different level of complexity are devoted to this problem for particular areas and weather conditions leading to classification of ozone formation into categories of chemical regime (Sillman, 1995). The detailed classification of ozone production regime helps to determine which of the two precursors must be targeted in ozone reduction strategy. Examples of such diagnose and of high-order sensitivity analysis are shown in Cohan, Hu and Russell (2004) and Cohan and Russell (2004).

The aim of this study is much more modest. It is to demonstrate the ozone formation sensitivity of CMAQ CB-4 chemical mechanism to variations in the main ozone precursors for a randomly selected period of time over specific geographical area. For the purpose, four emission scenarios are run with CMAQ. The basic scenario exploits the emissions described above (scenario **a**). The other three are: NO_x emissions reduced to 50% (scenario **b**), VOC emissions reduced to 50% (scenario **c**) and both NO_x and VOC emissions reduced twice simultaneously (scenario **d**).

The meteorological situation over the Balkan Peninsula for this period can shortly be described as follows: (a) the dominant wind was from North and North-East in Bulgaria and Romania and mainly from West for the remaining part of the domain; (b) the temperature was between 20 and 30 degrees Celsius, but the wind speed was high (especially at the end of the period) and (c) it was cloudy most of the time.

This shows that one should not expect very high ozone concentrations. This is confirmed by the results of calculations. Moreover, in the Western part of the region (where the wind was from the West) the ozone concentrations are higher.

As already said, the model was run for a period of 8 days (1-8 June, 2000) but only the last four days are commented, here. The graphical presentation of the results for only one of these days (7 June, 2000) is given in Fig. 3. The same trends are also observed for the other days in the period.

The NO₂ concentrations during all the time are below 3 ppb in the whole space domain excepting small regions around Athens and Istanbul, as well as (but in a smaller degree) in Bucharest. Some special behaviour of the results obtained with the NO_x scenarios could be expected in these regions. The results obtained by the scenario **b** (50% reduction of the NO_x emissions) confirm this. In the upper-right-graph in Fig. 3, the ratios of the ozone concentrations obtained by the scenario **b** and by the Basic scenario (scenario **a**) are given (in %). It is seen that the reduction of the NO_x emissions leads to a considerable reduction of the ozone concentrations in nearly the whole space domain. However, the ozone concentrations in the regions around Athens and Istanbul are increased. This effect is not pronounced for the region around Bucharest, where the NO₂ concentrations are not much higher.

The VOC scenario (scenario **c**) does not lead to great changes of the ozone concentrations. In the lower-left plot of Fig. 3, the ratios of the scenario **c** produced ozone concentrations and those obtained by the Basic scenarios are given. It is seen that the ozone concentrations in a large part of the domain are slightly increased when the VOC scenario is used.

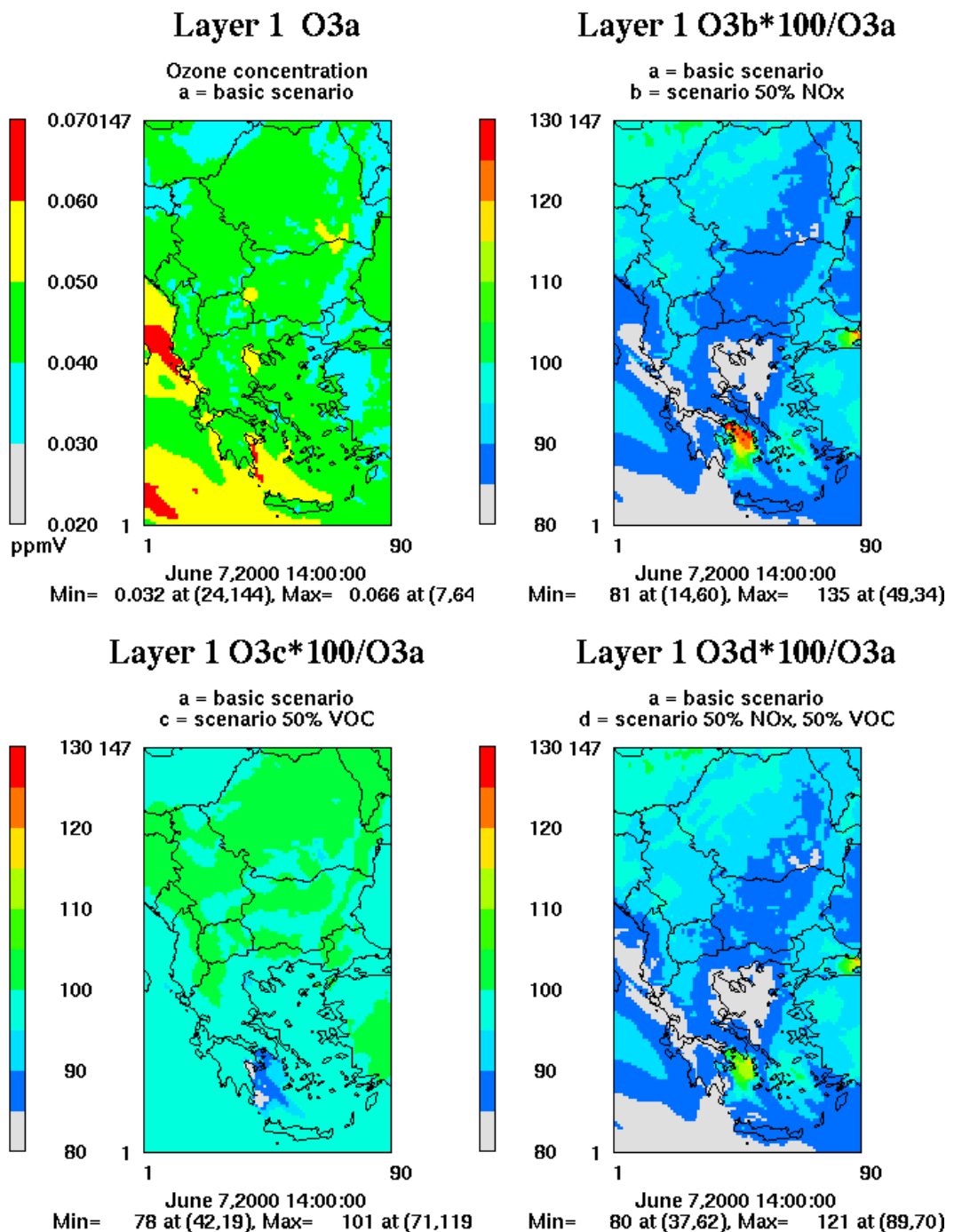
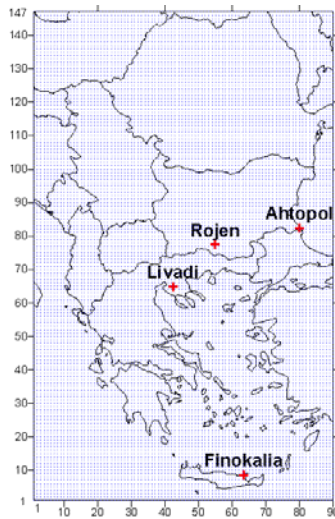


Fig. 3. CMAQ produced ozone fields for 7 June, 2000, 14:00 UTC:
up-left: ozone concentrations (Scenario **a**.); up-right: Scenario **b**/Scenario **a** in %;
down-left: Scenario **c**/Scenario **a** in %; down-right: Scenario **d**/Scenario **a** in %).

The results obtained by scenario **d** are compared with the results obtained by the Basic scenario on the lower-right plot of Fig. 3. It is seen that the results are quite similar to the results obtained when only the NO_x emissions are reduced. Two effects should be noted: (a) the areas where the reduction of only the NO₂ emissions leads to

an increase of the ozone concentrations are now reduced considerably and, moreover, the increased values are lower than the corresponding values in the in the upper-right plot and (b) the sizes of the areas where the reduction of the ozone concentrations is considerable (more than 10%) are slightly increased.

8. VERIFICATION AGAINST MEASUREMENT DATA



The number of background stations monitoring the ozone concentration is quite limited in the region. One can obtain only two such stations belonging to EMEP monitoring network that used to operate all the year 2000. These are the Greek stations GR02-Finokalia (35N30, 26E10, altitude 0 m) and GR03-Livadi (40N32, 23E15, altitude 850 m). It is worth to say that the coordinates of Finokalia in EMEP station list are wrong. The right values are (35N20, 25E40). Source of the data: <http://www.nilu.no/projects/ccc/emepdata.html>.

It occurs that during all the year 2000 two more stations were monitoring the ozone concentrations in Bulgaria in the frame of a research project (Donev et al., 1999, 2000). These stations are BG02-Rojen (41N40, 24E48, altitude 1700 m) and BG03-Ahtopol (41N58, 27E57, altitude 0 m). From these 4 stations two are at the sea shore, one is in-land (Halkidiki peninsula) and one - in the Rhodope mountain, peak Rozhen. Their location is shown in the picture on the left. Comparison of CMAQ calculated and DEM interpolated hourly ozone values with the measurements performed in these four stations for the period 5-8 June, 2000 can be seen in Fig. 4.

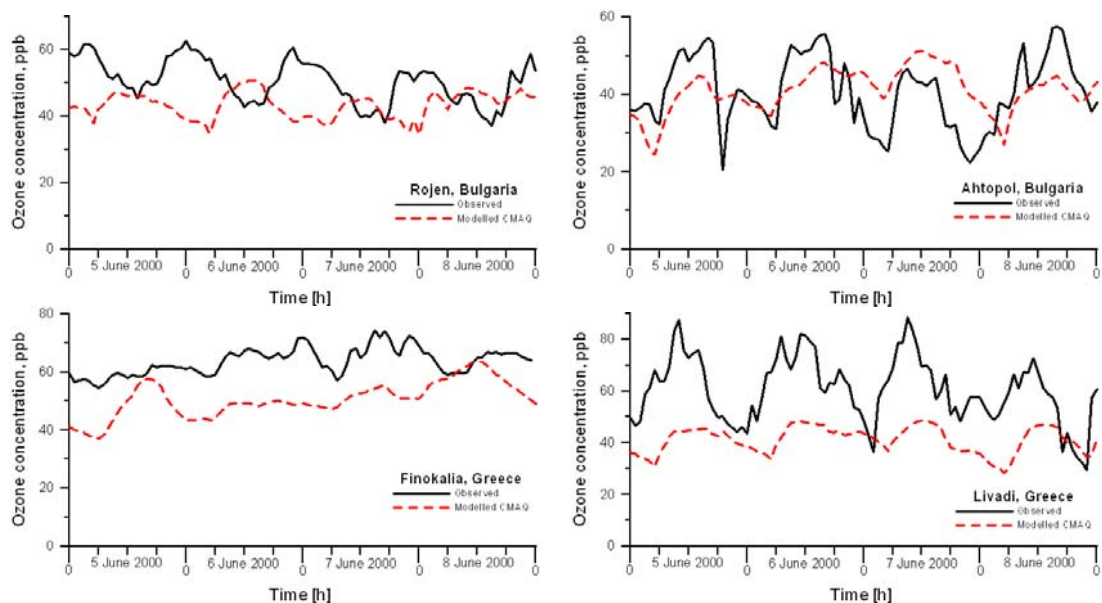


Fig.4. Comparison of CMAQ calculations with observations

The main features in Fig. 4 can be commented as follow:

- **Finokalia:** There are practically no diurnal variations of the measured ozone concentrations and of the CMAQ results.

- **Livadi and Ahtopol:** Both the model results and the measurements show the typical for the ozone concentrations diurnal cycles. The maxima and the minima of the model results are slightly shifted forward in relation to the corresponding minima and maxima of the measurements.

- **Rojen:** The measurement results at Rozhen show somewhat strange behaviour. They demonstrate opposite to the normal ozone diurnal cycle which, obviously, means that the ozone measured there is not produced in place but transported from some other regions. This possibly is the reason that stations with altitude over 1200 m are not usually used for comparisons with model data (Berge et al., 1994). The model produces maximal concentrations during the day and minimal concentrations during the night.

The main conclusion from Fig. 4 is that CMAQ-produced ozone concentrations underestimate the observed values all the time and in all stations. In the same time, the diurnal amplitudes are less than the observed ones. The basic reason for this is possibly the fact that the biogenic VOC emissions are not accounted for in the E_CMAQ calculations. These emissions would play considerable role in the ozone formation in this relatively warm region because of the intensive production of isoprene and other VOC with well expressed diurnal variations. One of the most important tasks in future investigations will be the attempt to include a reliable biogenic emission mechanism in CMAQ pre-processing software.

9. CONCLUSIONS

It can be concluded from these preliminary results that Models-3 air quality system describes quite reasonably the ozone formation in southeast Europe and it can be used for further complicated investigations of photo-oxidation regime on the base of specialized indexes determining the damages caused by high ozone levels in the region (as done in Zlatev and Syrakov, 2004b). For the purpose a lot of additional investigations and tests must be done in order to obtain realistic results to be used in emission trade negotiation process, as well as in decision making for elaboration of proper abatement strategies.

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