A SYSTEM FOR FORECAST OF BACKGROUND AIR POLLUTION LEVELS OVER BULGARIA: DESCRIPTION AND VALIDATION

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Abstract

Both, the current level of air pollution studies and social needs in the country, are in a stage mature enough for creating Bulgarian Chemical Weather Forecasting and Information System. For the purpose, a project is funded by the National Scientific Fund aiming at designing and setting up such a system able to provide timely, informative and reliable forecast products tailored to the needs of various users. The system is foreseen to provide in real time forecast of the spatial/temporal Air Quality behaviour for the country and (with higher resolution) for selected sub-regions and cities on the base of the weather forecast and national emission inventory. The system is aiming at integration within the European Chemical Weather Forecast community, utilizing knowledge and data, as well as the "model ensemble" possibilities.

The country-scale part of the system is designed and is being tested. It is based on the US EPA Models-3 System: MM5, CMAQ and SMOKE (partly). The meteorological pre-processor MM5 is drive by ALADIN output, ALADIN being the national numerical weather forecast tool. In this stage, the emission input exploits the high-resolution TNO emission inventory with tendency to make use of local data. The boundary conditions are prepared by a similar system running operationally in Aristotle University of Thessaloniki, Greece (AUTH). Special interface is created to retrieve in real time the AUTH-system forecasts producing boundary files uploaded to dedicated server in Bulgaria.
In the presentation, detailed description of the System's structure will be given together with first results of its testing.

Key words: *Air Pollution modelling, Chemical Weather forecast, Emission modelling*

1. INTRODUCTION

Air quality forecasting is extremely challenging scientific problem, which has recently emerged as an important priority in many urbanized and industrialized countries due to the increasing harmful effect on health and environment caused by airborne pollution constituents like ozone and particulate matter (PM). Elevated concentrations of air pollutants - especially during more extreme episodes - can pose a significant public health concern. According to WHO (2004), there is increasing evidence for adverse effects of air pollution on both the respiratory and the cardiovascular system as a result of both acute and chronic exposure. In particular, a significant reduction of life expectancy by a year or more is assumed to be linked to long-term exposure to high air concentrations of particulate matter (PM). One of the requirements of the Ozone Directive 2002/3/EC (European Parliament, 2002) and a key issue of the many European Air Quality Directives and programs is the availability of a trustworthy air pollution forecast system. The goals of reliable air quality forecasts are the efficient control and protection of population exposure as well as possible emission abatement measures. In last years the concept of "chemical weather" arises and in many countries respective forecast systems are being developed along with the usual meteorological weather forecasts (see, for instance, Sofiev at al., 2006, Poupkou et al., 2008a, 2008b, Monteiro et al., 2004, San Jose et al. 2004, 2006, 2007, and others).

As air pollution crosses national borders, it would be cost-effective and beneficial for citizens, society and decision-makers that national chemical weather forecast and information systems would be networked and seamless across Europe. For the purpose a new COST Action ES0602 "Towards a

European Network on Chemical Weather Forecasting and Information Systems (ENCWF)" (http://www.chemicalweather.eu/) was launched aiming at providing a forum for harmonizing, standardizing and benchmarking approaches and practices in data exchange and multi-model capabilities for air quality forecast and (near) real-time information systems in Europe. It is supposed to examine existing and work out new solutions for integrating the development efforts at national and international levels. It will serve as a platform for the information exchange between the meteorological services, environmental agencies, and international initiatives.

Bulgaria was one of the first countries that join COST Action ES0602, but so far it do not possess its own Chemical Weather Forecast system in spite air pollution causes serious damage on human health and constructions, especially in the urban and industry areas. The harmful effects of the background pollution on natural and agricultural ecosystems are noticeable, too. All this invokes a project financially supported by the National Science Fund with the Ministry of Education and Science aiming at creating Bulgarian Chemical Weather Forecast and Information System (BgCWFS) intended to provide timely, informative and reliable forecast products tailored to the needs of various users and decision-makers. The system is planned to have nesting structure, starting from the region of Bulgaria and nearest territories as a whole (background pollution) and zooming to smaller areas of interest.

In the paper, the structure of the country part of the system will be described. Some preliminary results addressed to model validation and verification will be presented, as well.

2. OPERATIONAL DESIGN OF BgCWFS

The country part of BgCWFS is designed in a way to fit the real-time constraints and to deliver forecasts twice a day (00 and 12 UTC) for the next 48 hours. US EPA Models-3 air quality modelling system is used, here, consisting of:

- CMAQ (Denis et al.,1996, Byun and Ching, 1999, Byun and Schere, 2006) - Community Multi-scale Air Quality model (http://www.cmaq-model.org/) being the chemical-transport model (CTM) of the System;
- MM5 (Dudhia, 1993, Grell et al., 1994) - the 5th generation PSU/NCAR Meso-meteorological Model (http://box.mmm.ucar.edu/mm5/) used as meteorological pre-processor to CMAQ, and
- SMOKE (Coats and Houyoux, 1996, Houyoux and Vukovich, 1999, CEP, 2003) - Sparse Matrix Operator Kernel Emissions Modelling System (http://www.smoke-model.org/) being the emission pre-processor to CMAQ.

Meteorological forecasts are obtained at the main synoptic terms using the MM5 mesoscale meteorological model forced by ALADIN output, ALADIN being the national weather forecast tool. Three nested modeling domains are set: ALADIN domain being the outer and the biggest one. MM5 domain is nested in it and CMAQ domain, covering Bulgaria, is nested in the MM5's one. The ALADIN meteorological forecast is carried out with a spatial resolution of about 12 km, while MM5's (respectively CMAQ's) resolution is 10 km. The MM5 vertical structure consists of 23 σ-levels with varying thickness, extending up to 100 hPa height. Proper physical options are set. The FDDA option of MM5 (Stauffer and Seaman, 1990) is **switched on** keeping its forecast close to the ALADIN one. MM5 starts its calculation 12 hours earlier for spin-up reasons (60-hour run).

In Fig. 1, the data flow diagram for one 48-hour cycle is displayed. In the boxes, together with the names of the system's elements, the format of the respective output is given. The white boxes present Models-3 components, the brown ones – the created interface modules (FORTRAN codes). The green boxes present the data input to the system. First of all, this is the meteorological forecast created by ALADIN which calculations drive MM5. MCIP, the Meteorology-Chemistry Interface Processor, is part of CMAQ and together with the needed meteorological parameters prepares some other data (fluxes, dry deposition velocities etc.) to be used by CMAQ and SMOKE. Area Source (AS) gridded inventory feeds the AEmis (AS emission processor). The Large Point Source (LPS) inventory is input to SMOKE, together with the ambient meteorological data as to produce LPS emissions (LPS processor). The met-data together with the gridded land-use are used by SMOKE to produce biogenic emissions (BgS processor). SMOKE is used once more to merge these 3 emission files in a model-ready emission input.

Apart these two main inputs – meteorology and emissions – CMAQ needs initial and boundary conditions. The initial conditions are taken from previous CMAQ run. The case with the boundary conditions (BC) is much more complicated. BCs are of great importance for small regions like Bulgaria, as it will be shown later. For such small areas, multiple nesting is usually applied, starting from big regions (continent, hemisphere). Constant climatic pollutants' values are set at these domains boundaries. Each next nested domain takes its boundary conditions from the pollution concentration already calculated over the bigger one. All this is related with many complications as necessity of respective meteorological forecast and emission data, needs of powerful computing facilities and large computational time.

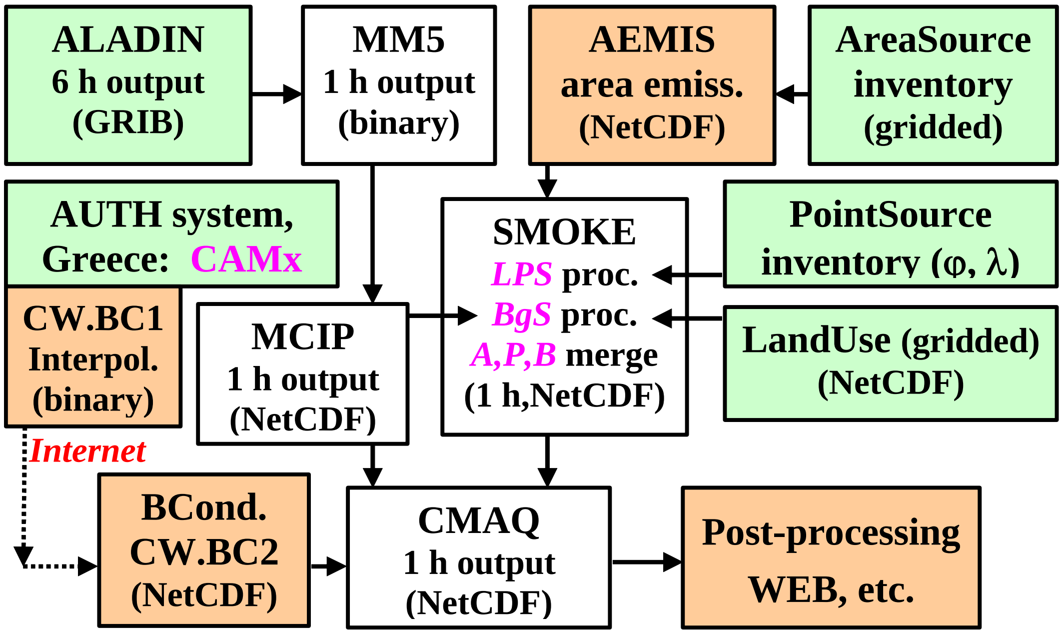

Fig. 1. 48-hour data flow of CW forecast calculations.

In BgCWFS, the boundary conditions are provided by the chemical weather forecast system running operationally in Aristotle University of Thessaloniki (AUTH), Greece (Poupkou et al., 2008a, 2008b). AUTH system exploits the nested domain approach, just described. The air quality forecast is carried out for Europe (50 km spatial resolution), the Balkans (10km) and Athens (2km) using a modelling system which consists of the prognostic meteorological model MM5 and the photochemical air quality model CAMx (ENVIRON, 2006). Annual anthropogenic emission data were provided by TNO. The emission data were temporally disaggregated in order to support the photochemical simulations. A biogenic emission model has been integrated in the forecast system. Over the European region MM5 is forced by AVN/NCEP global forecast. AUTH system is run once a day producing 3-day pollution forecast.

The 3-day real time boundary conditions for CMAQ are prepared in two steps. First, CAMx data is interpolated for CMAQ boundary points, an operational procedure that takes place in AUTH, results being uploaded to a dedicated sever in Sofia. Here, this data is processed on-line as to produce 3-day CMAQ-ready boundary condition file (see Fig. 2). Usually, there is a delay of AUTH data delivery according to the BGWC run. But the fact, that the AUTH forecast is 3-day long permits to overcome this obstacle: for BgCWFS's 00:00 run the previous boundary file (prepared from the previous day AUTH forecast) is used. For the second run at 12:00 the new BC file is already issued and can be used by the system.

3. EMISSION MODELLING

CMAQ demands its emission input in specific format reflecting the time evolution of all polluters accounted for the used chemical mechanism. The emission inventory usually is made on annual basis for, as a rule, big territories (municipalities, counties, countries etc.) and many pollutants are estimated as groups like VOC and PM2.5. In preparing CMAQ emission file, a number of specific estimates must be done. First, all this information must be gridded. Secondly, time variation profiles must be over-posed on these annual values to account for seasonal, weekly and daily variations. Finally, VOC and PM2.5 emission estimates must be split into more defined compounds (**speciation**) in order to be properly modeled for chemical transformations and deposition. Obviously, models are needed as emission pre-processors to CTMs. Such a component in EPA Models-3 system is SMOKE. Unfortunately, it is highly adapted to the US conditions and is partly used, here, only for calculating LPS and BgS emissions and to merge AS-, LPS- and BgS-files into a CMAQ emission input file.

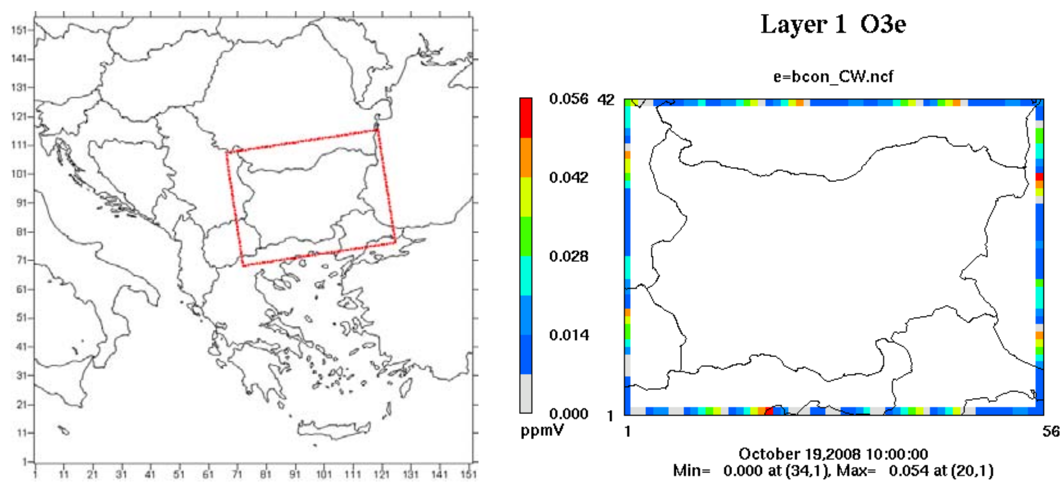

Fig. 2. Second domain of AUTH/CWFS with CMAQ boundaries (left) and CMAQ-ready BC file for BgCWFS (right)

As shown in the data flow diagram in Fig. 1, the AS anthropogenic emission file is prepared by interface programs AEmis. Input to these interfaces is gridded emission inventories for year 2000. For the moment, TNO inventory (Visschedijk et al., 2007) is exploited. This inventory's resolution is 0.25°×0.125° longitude-latitude, that is on average 15×15 km. GIS technology is applied to redistribute emissions from this data base over BGWC's grid, results transformed in AEmis input file.. It must be mentioned that the TNO inventory is elaborated for AS and LPS separately, distributed over 10 SNAPs (Selected Nomenclature for Air Pollution) classifying pollution sources according the processes leading to harmful material release to the atmosphere (EMEP/CORINAIR, 2002). The inventory contains 8 pollutants: CH4, CO, NH3, NMVOC (VOC), NOx, SOx, PM10 and PM2.5.

The temporal allocation is made on the base of daily, weekly and monthly profiles, provided by Builtjes et al. (2003). The temporal profiles are country-, pollutant- and SNAP-specific. The speciation procedure is dependent on the Chemical Mechanism (CM) used. CMAQ supports different CMs. Here, the Carbon Bond, v.4 (CB4) is exploited (Gery et al., 1989). In the used Version 4.6 of CMAQ the CB4 is upgraded with the Version 1.7 of ISORROPIA aerosol model (Nenes et al., 1998). It requires splitting of VOC to 10 lump pollutants (ISOP, OLE, PAR, ALD2, TERPB, XYL, ETH, NR, FORM, TOL) and PM2.5 to 5 groups of aerosol (PSO4, PNO3, POA, PEC, PMFINE).

A specific approach for obtaining speciation profiles is used here. The USA EPA data base (http://www.epa.gov/ttn/chief/emch/speciation/) is intensively exploited. A Bulgarian emission expert has found coincidence between main Bulgarian sources for each SNAP with similar source types from US EPA nomenclature. The weighted averages of the respective speciation profiles are accepted as SNAP-specific splitting factors, weights being the percentage of contribution of each source type in total Bulgarian emission in particular SNAP. In such a way VOC and PM2.5 speciation profiles are derived. It must be noticed that the choice of source types and their contribution to the respective SNAP emissions are country specific, i.e. the obtained speciation profiles are applicable for Bulgarian territory, mainly.

As far as **gridded** emission inventories are input to AEmis, the program performs only speciation and time allocation for each grid cell and for each SNAP. The obtained hourly values of all processed pollutants are output as 2D NetCDF-file. SMOKE is used to produce LPS emission file. For the purpose, LPS inventory (coordinates, stack parameters, emissions) is transformed in required IDA format. Serious update of SMOKE's profiles' and references' files had to be made as to urge the model to produce Europe-specific results.

SMOKE currently supports BEIS (Biogenic Emissions Inventory System) mechanism, versions 3.13 (Schwede et al., 2005). The model is fed with gridded land-use data. It computes the normalized emissions for each grid cell and land-use category. The final step is adjusting the normalized emissions based on gridded, hourly meteorology data as produced by MCIP and output a model-ready biogenic emissions file. For preparing the gridded land-use file GIS technology is applied to USGS (US Geological Survey) data base with resolution 1 km. The output NetCDF file contains the hourly variations of CO, NO and the 10 VOC's lump pollutants (including Isoprene and Monoterpenes).

4. CMAQ CALCULATIONS AND POST-PROCESSING

Fourteen σ-levels with varying thickness determine the vertical structure of CMAQ. The Planetary Boundary Layer is presented by 8 of these levels. Besides meteorology and emissions, CMAQ needs also Initial Condition (IC) and Boundary Conditions (BC) input. As the CMAQ calculations are twice a day, the forecast for the respective hour of the day is IC for the next run. For the first day of operation of the system, climatic values are exploited as IC. The made errors decrease quickly with time because they are "blown" fast from such small territory. As to the boundary conditions, their handling was already described.

The daily CMAQ, v.4.6., output is a NetCDF file with 3D hourly data for 78 pollutants, from which: 52 gaseous, 21 aerosols (Aitken and accumulation modes), 5 aerosol distributions (3 by number, 2 by aerosol area).

The last box in Fig. 1 tags the post-processing activities that is quite important as to BgCWFS results become visual. First of all the post-processing program XtrCON_CW extracts part of the pollutants for archiving and further handling. Only surface values of the most important pollutants are saved. Saved on hourly basis are the next 17 pollutants:

- NO2, NO, HNO3, H2O2, O3, CO, SO2, NH3 (gases)
- PSO4, PNH4, PNO3, POA, PEC (aerosol)
- SOAA, SOAB (Anthropogenic and Biogenic secondary organic aerosol)
- FPRM, CPRM (fine and coarse PM).

These pollutants are more or less monitored and part of them is referred in the legislations with the respective thresholds. It must be mentioned that the sum of all aerosol compounds forms PM10 (usually measured) and PM2.5=PM10-CPRM. PM10 and PM2.5 are added for archiving and further processing to the pollutants, just mentioned. All this data is stored in a single NetCDF file with the current Julian date and hour as file-name.

As to make the results of BgCWFS operation public, a specialized web site was created on a dedicated server of NIMH. For the moment, it presents the forecast for only 4 most important pollutants but plans for creating a more informative site exist. The preparation of respective graphs is included into the operational schedule of the System. After finishing the routine calculations, a special script is invoked creating the plots. PAVE v.2.3 is exploited for the purpose (http://paved.sourceforge.net/pave_doc/Pave.html). On the left side of the web-page, list of selected pollutants is given together with respective kind of plots. The 48-hour forecast can be displayed for each pollutant. It can be retrieved by putting the courser on the respective line in the forecast time scale at the right side of the graph. Putting the courser over Play-line invokes animation of the forecast. Under the graph, respective thresholds according to the local legislation are presented. For Ozone, 4 additional graphs can be invoked: daily maxima and daily 8-hour running averaged maxima for the first and for the second forecast days.

5. BgCWFS VALIDATION AND VERIFICATION

Before starting the use of BgCWFS, validation of model results is needed in order to assure the quality and credibility of delivered output. Here, validation for ozone and for 2000 is done. The reasons are two. Firstly, the up-to-date national emission inventory is in a stage of completion and is not available. Secondly, the background measurements in Bulgaria are quite scanty. Using the 2000 TNO inventory, respective meteorology and measurement data are necessary. Fortunately, in the frame of research project (Donev et al., 2002), hourly measurements of ozone for all year 2000 have been performed in two background points of the country – peak Rojen (Rodopi mountains, south Bulgaria) and Ahtopol (small Black sea town, southeastern Bulgaria). Part of the validation results will be presented, here.

Four scenarios are calculated as to distinguish the importance of different processes to ozone formation. Scenario A is basic – all kind of emissions and boundary conditions are accounted for. At scenario B the boundary values of all pollutants are set to zero. Scenarios C and D are as A and B, respectively, but biogenic emissions are excluded. Further on, results only for Rojen will be presented and discussed. Ahtopol's results are quite similar.

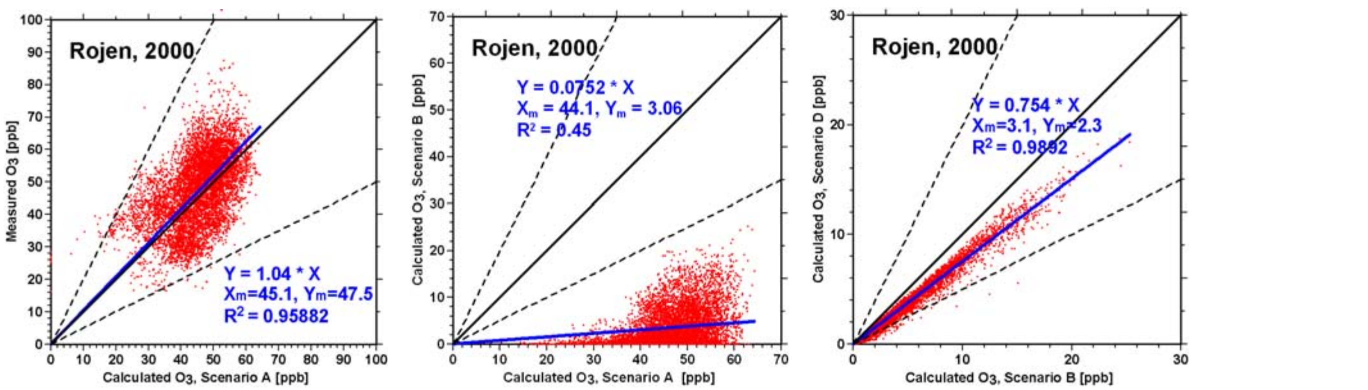

Fig. 3. Scatter diagrams: Measurements *vs.* Scenario A (left); A *vs.* B (middle), B *vs.* D (right)

In the left graph of Fig. 3, one can notice that almost all scatter points are into FA2 boundaries. The fitting lines are quite close to the ideal fitting line showing slight underestimation of measurement data. The correlation coefficients of both fits are quite high that reflects the good quality of the simulation. The comparison of scenarios A and B (graph in the middle) shows how important is the correct accounting for boundary conditions, especially at air quality calculations over such a small domain. BC form almost 90 % of pollution levels in the region. The space structure of respective fields is quite different – in case B, maximal ozone values are concentrated in the middle of the domain while in opposite case they ollow the main pollution sources.

The scatter diagram scenario A *vs.* scenario C (biogenic emissions excluded) is not shown in the Figure because the scatter points lay very close to the ideal fitting line, correlation coefficient close to 1. In contrary, the right graphs on Fig. 3 (case with zero fluxes through the boundaries) show, firstly, ozone values at least three times lower than in the opposite cases. Secondly, in this case the biogenic emissions are very important – the ozone values become 30% higher when this emission source is switched on. These effects can be explained with the help of the Empirical Kinetic Modeling Approach diagram (Finlayson-Pitts and Pitts, 1986). Obviously, in case of absence of powerful NOx sources in the vicinities of both measuring points, the ratio NOx/NMVOC gets in the so-called "NOx-limitation" regime – ozone production is not sensible to VOC changes. In the opposite case (right graph in Fig. 2), when no "foreign" pollution enters the computational domain, NMVOC/NOx<8 and the ozone production is sensible to changes of VOC concentration.

According to European ozone directive (European Parliament, 2002) several indexes, mainly related with exposure, are more important than ozone concentrations. Such are AOT40 (Accumulated Over Threshold of 40 ppb), NOD60 (Number Of Days with 8-hour average greater than 60 ppb) and others. They were calculated from both measurement and scenario A time series. It must be noted that the model underestimate these indexes in both stations. The possible reason is that the model maxima are smaller as a whole.

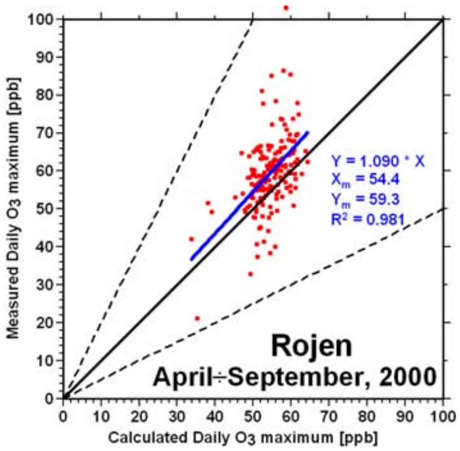

Fig. 4. Daily maxima scatter diagram

As far as the best way to compare ozone data is the usage of daily maxima, the simulation quality of BgCWFS system is demonstrated once more in Fig. 4, where the scatter diagrams of measured *vs.* calculated daily maxima (for Rojen, again) is displayed. One can notice that in spite the fitting line are close to the ideal one, correlation coefficients being over 0.95, measured daily maxima are somewhat underestimated. The above mentioned indexes are quite sensible to the ozone maxima and this explains the failure of the model to reproduce correctly the measured indexes. Nevertheless, the simulation results are rather good. This means that BgCWFS system is able to reproduce quite realistically the each day ozone variations and serve as air quality forecast and information system.

The summertime average ozone concentrations and average daily maxima (ADM) fields can be seen in Fig. 5. One can notice some resemblance between space patterns but the values are quite different – the maximum of climatic ozone field is close to the minimum of the ADM-field. The common feature is the high values over the Black sea that is trivial fact – the dry deposition of ozone is practically zero, there. The second common feature is the maximum in the south-western part of the country, trail from Sofia and other highly industrialized areas near around. Additional maximum can be noticed in ADM field. It is in the southern part of the country where 3 lignite burning thermal power plants are situated. This maximum is much better seen in averaged SO2 and PM fields.

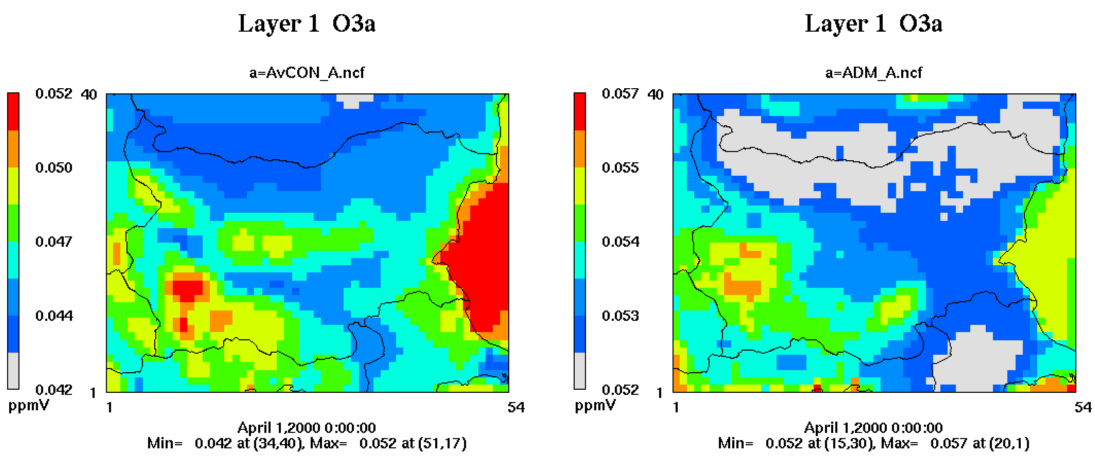

Fig. 5. Summertime average ozone concentrations (left) and average daily maxima (right)

6. CONCLUSION

Evaluation of BgCWFS simulations showed that the modelling system has a satisfactory performance with respect to O3 as shown from the different plots discussed. Despite using boundary conditions from another modelling system the basic spatial and temporal O3 patterns are captured by the model. The best simulation quality refers summer time daily maximums. There are essential discrepancies when estimating the O3 indexes recommended by EU Ozone Directive. The reasonable performance of the BgCWFS for the past time simulations justifies its use for future forecast and information to be used of various users.

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