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CMAQ (COMMUNITY MULTI-SCALE AIR QUALITY) ATMOSPHERIC DISPERSION MODEL ADAPTATION FOR HUNGARY

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Abstract: Up to our days it has become more important to measure and predict the concentration of atmospheric pollutants – harmful contaminants such as dust, aerosol particles of different sizes, nitrogen compounds, and ozone. The Weather Research and Forecasting (WRF) model has been applied at the Department of Meteorology at Eötvös Loránd University for several years now. This model is suitable for weather forecasting purpose and may also provide input data for various environmental simulation softwares (e.g. DNDC, AERMOD). By adapting the CMAQ (Community Multi-scale Air Quality) model we have implemented a coupled air-quality – meteorological environmental model system, primarily for the representation of atmospheric ozone. The modular structure of the CMAQ allows successful and fast simulations with different scales from global to local. In our present investigation it is important to apply different scale emission databases and describe the initial distribution of pollutants using a background model. We are going to adapt CMAQ model to Hungary. The meteorological parameters are the primary physical forces in the atmosphere. We used WRF model in order to generate the meteorological driver database and the so-called SMOKE model for the generation of the input emission database. WRF/CMAQ model system has been run on a three-level one-way nested grid of 108/36/12 km grid spacing, covering Central Europe, the Carpathian Basin and Hungary, respectively. We used the CMAQ 5.0.1 version which includes i) an updated version of the carbon bond "CB05" gas-phase mechanism (with active chlorine chemistry and updated toluene mechanism), ii) sixth-generation aerosol mechanism (with sea salt and specialized PM among others), iii) Cloud module, etc. For better quality simulations we used the Geos-Chem model results as initial and boundary conditions. We studied ozone forecasts for Hungary based on different model settings and transition time using several verification methods. This paper presents the outline of the project work and the first results of concentration calculations compared to the national ambient air stations data.

Key words: *air-quality modelling, ozone, CMAQ, SMOKE, WRF, emission, model adaptation*

INTRODUCTION

In discussions about the forecast of ozone concentrations e.g. during summer smog episodes, relative importance of horizontal advection, vertical mixing, and chemical production is still an unsolved problem. The solution of this problem probably lies in sensitivity studies with complex numerical models including both meteorology and chemistry (Neu et al., 1994). The forecasts of ozone concentration are important by reason of the harmful effects of ozone (O_3) on both human health and the environment (McDonnell et al., 2002; Colette et al., 2012). Breathing ozone may trigger several health problems including chest pain, coughing, throat irritation, and congestion (Gryparis, 2004; Amann et al., 2011). It may worsen bronchitis, emphysema, and asthma (McDonnell et al., 2002; Holicska, 2008). Ground-level ozone also may reduce lung function and inflame the linings of the lungs. Repeated exposure may permanently scar lung tissue. In the US the human exposure to high concentration of ground level ozone continues to bother many areas in spite of the implementation of government-mandated emissions control strategies (Finlayson et al., 2000). This is because the control of ground level ozone is more difficult than for many other primary pollutants because ozone is a secondary pollutant. In case of other primary pollutants, a reduction in emissions results approximately proportional reduction of pollutants. However, as a secondary pollutant that is formed from primary pollutants and other chemical species in the atmosphere, ozone does not necessarily respond in a proportional manner to reductions in precursor emissions. Air quality modelling provides a good alternative to study the physical and chemical mechanism of ozone formation because modelling can provide good temporal and spatial resolution for a

wide variety of pollutants (Bozó, 2005; Leelőssy et al., 2014). For instance, regional air quality models such as the US EPA's Community Multiscale Air Quality model (CMAQ/Models-3) can be used to generate hourly ambient concentration fields for ozone, PM10, PM2.5, and many other pollutants, which allows researchers to study the relationship between pollution and health outcomes for times, locations, and pollutants for which monitoring data are not available (Lu et al., 2008; Pay et al., 2010).

In this work, an air quality modelling system was applied to PhD thesis to adaptation and study ozone concentration and relationship between other chemical substances (eg. NOx, VOCs, HAPs) for Hungary. Some studies have been conducted to investigate ozone and other pollutant emission concentration with different areas, model domain and chemical mechanism. In USA the model system has been configure and studied up-to-date, e.g. Wong et al. (2012) studied WRF – CMAQ model with two-way coupled system with some pollutant (ozone, PM10, PM2.5). They concluded that ozone has absorption bands in the long wave radiation bands and can thus absorb outgoing radiation. Efforts are underway to implement ozone feedback in the coupled WRF-CMAQ system and study the impact of ozone on long wave radiation using the RRTMG long wave radiation scheme. Boulton et al. (2012) studied the 2012 year emissions in Toronto. This was a multi-year project, which has been concluded with assessing behaviour of the model for ozone and PM2.5. In Europe WRF-CMAQ model system has been applied for research goals in many institutes e.g. in Scotland (Pederzoli, 2008) and in Bulgaria with cb04 mechanism and on 14 vertical levels (Syrakov et al., 2015).

In this article we represent the model attributes and the selected case of our studies, then we examine the first model forecast results and conclude the attribute of the model. The main goal is we understand the model attitude in the area of the Europe and Carpathian Basin.

CASE SELECTED AND MODEL CONFIGURATION

In this chapter the model inputs, the weather situation for the case simulation, selected chemical mechanisms and design of numerical simulations will be presented.

We used 3 different model input for the applied model system, i) the meteorology input has been utilised by WRF model outputs and MCIP (Meteorology-Chemistry Interface Processor) to create netCDF-formatted input meteorology files that are used by the emissions model (SMOKE) that computes emissions inputs to CMAQ. ii) For the emission database we applied EMEP (European Monitoring and Evaluation Programme) ASCII files and recalculate with SMOKE for the CMAQ emissions, iii) for construction of initial and boundary conditions we run GEOS (Goddard Earth Observing System) – Chem model system and used PseudoNetCDF python script. The model system settings was Single-moment 3-class in WRF model, Nei2005 (National Emissions Inventory) CB05 with SOA (secondary organic aerosol) in SMOKE model, tropospheric chemistry mechanisms (aka "Full-chemistry") with 47 levels in GEOS-Chem model system. In SMOKE model we used this chemical mechanism, for the reason that we could use and compare 2 different chemical mechanism in CMAQ model, as Carbon Bond version 5 chemical mechanism (cb05tucl_ae6_aq, TUCL (toluene and chlorine mechanism)) and explicit air toxics chemistry (cb05tump_ae6_aq, TUMP (Multi-pollutant mechanism)).

The CB05 mechanism includes updates in toluene chemistry, in homogeneous hydrolysis rate constants for dinitrogen pentoxide (N_2O_5), and in chlorine chemistry. Whitten et al. (2010) developed new condensed toluene chemistry for the CB05 mechanism. The International Union of Pure and Applied Chemistry (IUPAC) now suggest using only the bimolecular homogeneous hydrolysis of N_2O_5 and also recommend a lower rate constant for the reaction. The existing chlorine chemistry contains 21 reactions involving chlorine. The cb05tump_ae6_aq mechanism predicts criteria air pollutants and several hazardous (toxic) air pollutants based on the 5.1 version of the cb05tucl_ae6_aq mechanism. The cb05tump_ae6_aq mechanism modifies the cb05tucl_ae6_aq mechanism (Sarwar et al., 2008; Whitten et al., 2010) to predict several Hazardous Air Pollutants (HAPs). It accomplishes the goal by adding mercury compounds, acrolein, 1,3-butadiene and reactive tracers to the cb05tucl_ae6_aq mechanism. The first three HAPs require adding complex chemical kinetics to the original photochemical mechanism. The reactions involving mercury do not alter predictions from the cb05tucl_ae6_aq mechanism because elemental mercury is treated as a reactive tracer with inert daughter products.

In our case study we selected an anticyclone weather situation in Europe at 3th decade of September 2012. The weather of the Carpathian Basin had been developed by the above-mentioned anticyclone, which was placed to the east on Saturday (22th September 2012), and faintly more humid air had been flown over Hungary. In the verification study we used several measurements from air-quality monitoring stations in 4 different countries (Austria, Hungary, Romania, and Slovakia). We selected Győr (47°40'40.8"N 17°39'26.6"E), Budapest (47°28'33.0"N 19°02'24.8"E) and K-puszta (46°58'00.0"N 19°35'00.0"E) in Hungary, Ilmitz (47°46'00.0"N 16°46'00.0"E) and Masenberg (47°20'53.0"N 15°52'56.0"E), in Austria. We used data from Poiana Stampei (47°19'29.2"N 25°08'04.8"E) in Romania and from Chopok (48°56'00.0"N 19°35'00.0"E) and Topolniky (47°57'36.0"N 17°51'38.0"E) in Slovakia. All observation points (except Győr and Budapest) are EMEP stations; the databases are free of charge on the official EMEP website (http://emep.int).

As it was mentioned in the abstract earlier 3 model domains covering Europe, the Carpathian Basin and Hungary separately using one-way nested grid with horizontal resolution of 108, 36 and 12 km (Figure 1). The 36 km model domain covers the Carpathian Basin, and the 12-km domain covers Hungary. All nested domains have 43 vertical layers, and the model top is set at 50 hPa. The lowest 17 model sigma levels are among 1.0 and 0.80.

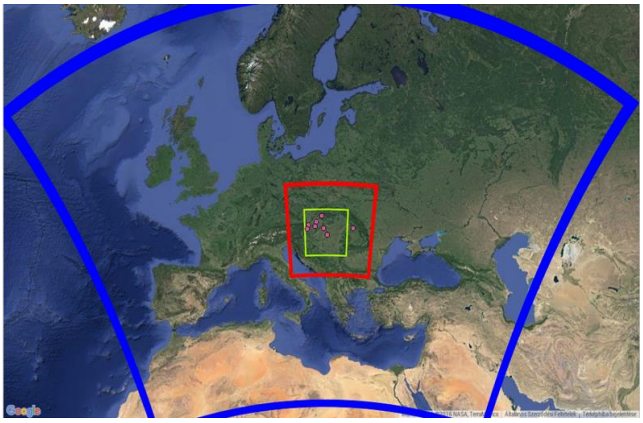

Figure 1. Nested domains employed by WRF – CMAQ – SMOKE model system, a) blue domain: 108 km grid (Europe), b) 36 km grid (Carpathian Basin), c) 12 km (Hungary), d) pink point: air-quality monitoring stations for vertification study

STRUCTURE OF THE MODEL SYSTEM

The CMAQ – WRF – SMOKE model system has a complex model structure (Figure 2). As it can be seen in the left side of the picture are the system models which gave the input files for the system. The WRF model makes the meteorological datasets with GFS data, which had to be modified to the system formats for SMOKE and CMAQ model. This task made the MCIP processor.

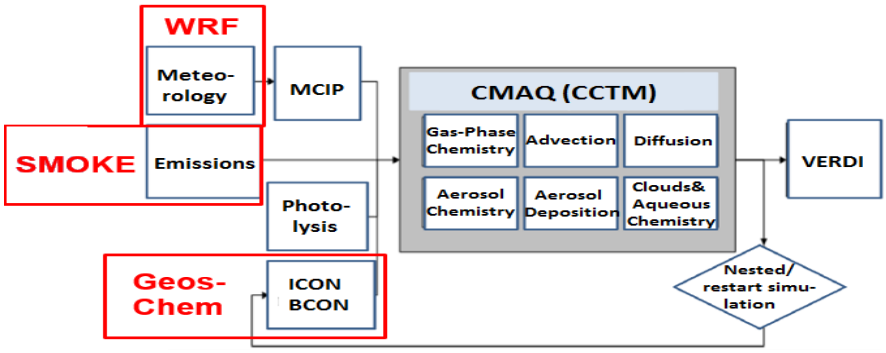

Figure 2. Sematic picture of the WRF – CMAQ – SMOKE model system. Black arrow shows the direction of the construction of the model system. Red box are optional model within the system, blue box is the sub processors in the CMAQ model

The emission dataset was constructed by SMOKE with meteorological data files, all emission sources (point, area, mobil or road) were in ASCII files. The boundary and initial concentration files were calculated by the GEOS-Chem model, which were put the CMAQ model inputs with PseudoNetCDF python scripts. After we had the input files for the CMAQ, we could run the forecast in our grid area. The outputs of the system were NetCDF files which we could visualise with VERDI program. The CMAQ, SMOKE models and VERDI program were made by CMAS centre (http://cmascenter.org), the applied GEOS-chem model is managed by the GEOS-Chem Support Team, based at Harvard University and Dalhousie University with support from the US NASA Earth Science Division and the Canadian National and Engineering Research Council (http://www.geos-chem.org/index.html). All models and programs are available free of charge. The model system is running at ELTE ATLASZ server.

CASE STUDY

In this section we represent the first results of our complex model for Hungary between 19th September 2012 and 27th September 2012. We would have liked to examine the model system quality with ozone datasets in the first place. Relationship between measurements and calculated concentrations had been studied, and after that the behaviour of each station with the modelled values was determined. First, we examined at 108 km grid how the model behaves in correlation with the data of the measurements. O_3 values were examined with both (TUCL and TUMP) chemical mechanisms thus we could decide which mechanism can be advantageous for forecasts of the Carpathian Basin.

In Figure 3. the correlation of measurements and forecasted concentrations were examined in K-puszta at 108 km grid. The model system was run with different start dates in order to estimate the time gap after that fairly good concentration results can be yielded from a 7-day-long forecast in comparison with the measurement data. Forasmuch as at 108 km grid the model system did not forecast the measured small concentration values at 21st, 22nd and 23rd, the values of 21st and 25th September 2012 were examined at 108 km, 36 km, 12 km grids in one point (K-puszta) separately. In the case of 25th September 2012 each the 108 km, 36 km and 12 km grid values were higher than the measurement data but morning and evening differences were smaller than forecast-measurement differences of 21st September 2012.

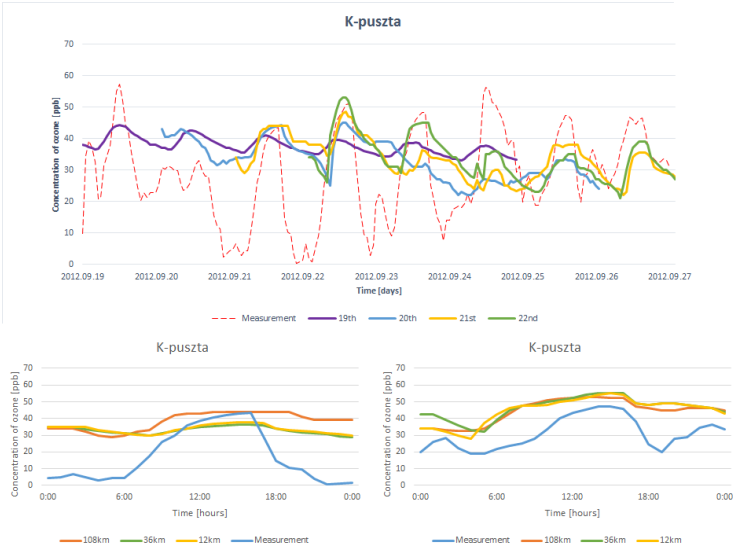

Figure 3. Concentration of ozone (O_3) on K-puszta. a.) eight-day ozone forecasts with different start time at 108 km grid (red:measurements, violet: 19th, blue: 20th, yellow: 21st, green: 22nd), b.) concentration of ozone at 21st September 2012. (blue: measurement, yellow: 12 km, green: 36 km, orange: 108km grid, 24-hour run) c.) concentration of ozone at 25th September 2012. (blue: measurement, yellow: 12 km, green: 36 km, orange: 108 km grid, 24-hour run)

Figure 4. shows the temporal changes of ozone concentration in the Carpathian Basin at 22nd September 2012. Spatial differences between the measurement data and the forecast values were studied. Figure 4. represents the above mentioned observation stations of the Carpathian Basin. The colour of each station

shall be interpreted according to the same ozone map scale. To study the temporal changes two stations (K-puszta and Chopok) were chosen where the results of the observations and the model system values were compared at 22nd September 2012. In the case of K-puszta the system values were higher than the measurement data. We assumed that these differences were due to initial concentrations presumably generated by an anticyclone weather situation. In the case of Chopok differences between the measurement data and the model values were smaller, presumably due to the location of the observation station (hilly region in a national park).

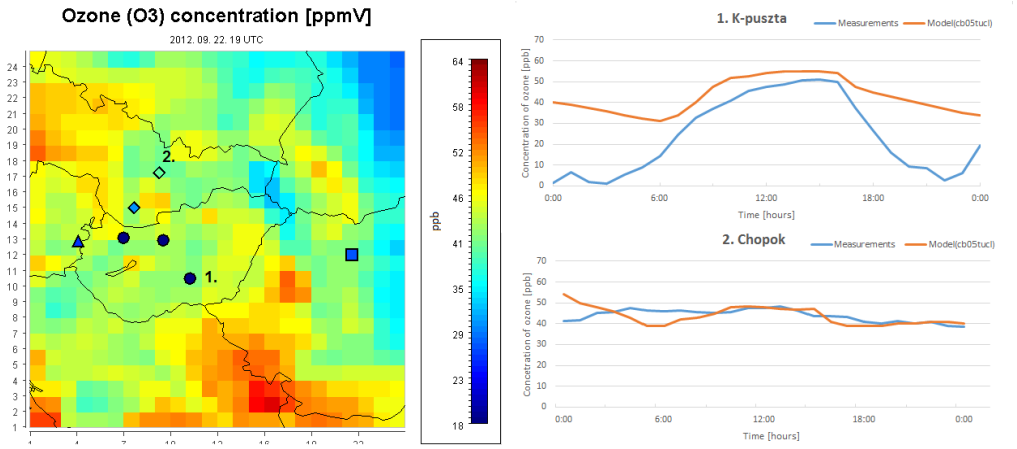

Figure 4. Ozone concentration forecast in the Carpathian Basin [ppb] a.) concentration values separately marked at the observation stations (19 UTC) (circle: Hungarian stations; triangle: Austrian stations; diamond: Slovakian stations; square: Romanian station) b.) measurements (blue line) and forecast values (orange line) for K-puszta (Dot 1.) and Chopok (Dot 2.) at 22th September 2012.

As it was mentioned above, in the Fig 5. the differences of the cb05 tucl and cb05tump chemical mechanism are shown with different visualisation methods. On the map of the spatial differences (a) there are chiefly small negative values, but in Romania and on the Great Plain (mostly in Eastern Hungary) higher positive difference values may appear. On the temporal change plot (b) and on the scatter plot (c) can be noticed that differences are particulary typical in the afternoon.

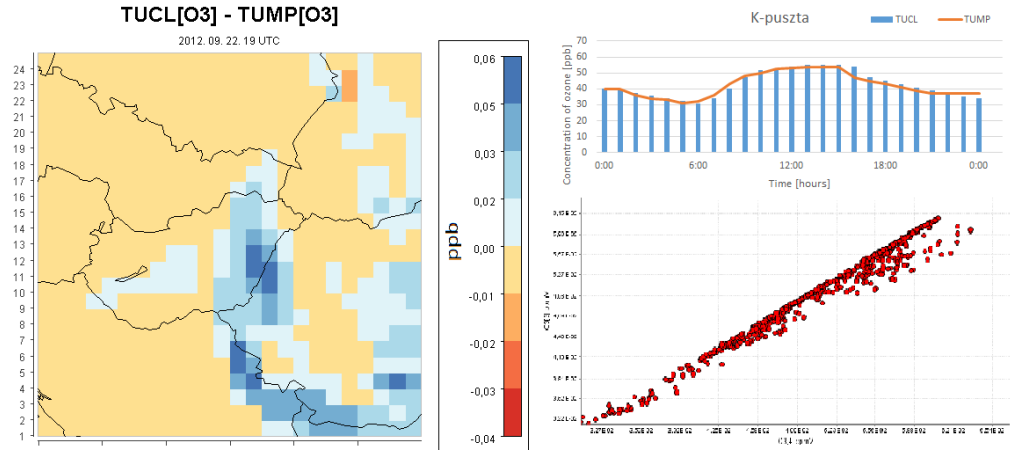

Figure 5. Differences of the two chemical mechanism ozone forecasts in the Carpathian Basin at 22th September 2012 at 36 km grid. a.) differences of ozone forecasts shown on map at 17 UTC 22nd September 2012 (brown: negative; beige and blue: positive) [ppb], b.) temporal change of O_3 concentration forecasted by two chemical mechanism on a one-day interval (orange: TUMP, blue: TUCL) [ppb], c.) scatter plot of the ozone concentration [ppmV] (x: TUCL, y: TUMP)

CONCLUSION

In this paper we introduced our adaptation of CMAQ – SMOKE – WRF model system on Europe and the Carpathian Basin in forecasting ozone concentration. After comparing the forecast values with the observation values we have concluded that a.) the model system shall be run about 3 days before fairly good forecast results can be yielded, b.) the forecast values depend on the initial and boundary values of the O_3 concentration, c.) the cb05 TUMP chemical mechanism produces typically lower forecast values than cb05 TUCL mechanism in the afternoon.

Our further plan is i) to configure a more detailed emission dataset for Europe and Hungary, ii) to examine the air pollution concentration forecasts for a full-year period, iii) to test the model sensitivity with other air-quality models (e.g. WRF-Chem) and iv) to build up an ensemble ozone forecast.

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