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NUMERICAL PROGNOSIS OF DISPERSION AND TRANSPORT OF POLLUTANTS IN ROMANIA, BASED ON EMISSIONS OF POLLUTANTS

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Abstract. Air pollution is one of the major problems of mankind, transport of pollutants extending far beyond the borders of the countries where they were produced, causing unpredictable, direct and indirect changes of the environment. The main tool for the study of this phenomenon consists of mathematical modeling of complex physical and chemical phenomena involved. In practice, air emissions are estimated on basis of measurements taken from selected sources being representative of the major categories and types. At national level, the Air Quality Evaluation Center (CECA) provides regular reports to the European Environment Agency (EEA) or the European Commission as requirements of Romania's lawful duties in air quality domain. The registry of emissions TNO/ MACC (Netherlands Organisation for Applied Scientific Research/ Monitoring Atmospheric Composition and Climate) contains emissions inventories which have been homogenized and checked in advance and obtained from emissions officially reported at sectoral level for each country. In this study, for the analysis of the weather numerical dispersion and transport of pollutants, it has been used the numerical air quality model WRF-CHEM version 3.5, centered over Romania, at the spatial resolution of 10 km, using as input data the TNO emission database for 2009. By interpolating values from the regular grid of the TNO database with the WRF-CHEM model 3.5 grid, monthly average values were obtained for each day of the week, for any parameter considered. Preliminary results obtained for different pollutants (for example: PM10, O_3) confirm the need to validate these results by implementing and integrating air quality forecasting model by assimilating different types of measurements (data model, gravimetric data observations, etc.).

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1. Introduction

Air quality is of major importance for the environment. Pollutants in large quantities can cause severe health problems, can destroy crops and are one of the factors responsible for global warming (EEA, 2010). The amount of pollutants emitted into the air we breathe has been greatly reduced when the European Union (EU) introduced policies and measures on enforcing air quality standards in the 1970s. The main legislative acts that set boundaries across Europe include the 2008 *Air Quality and Cleaner Air Directive 2008/50/ EC* and the 1996 *Framework Directive on ambient air quality assessment and management (96/62/ EC)*. Atmospheric pollution is one of the main problems of humanity, transport pollutants beyond the borders of the countries where they were produced, causing unpredictable changes, direct and indirect environmental impacts (WHO, 2003). The main tool of study of this complex phenomenon is represented by the mathematical modeling of the physical and chemical phenomena involved. In recent years, several models of atmospheric chemistry and transport (CTM) have been developed to better understand the physical and chemical processes of suspended particulate matter and they were also applied for operational air quality forecasts.

Particulate matter (PM) represents a mixture of solid particles and liquid droplets found in the air and can be emitted from a source (e.g. construction sites, fires, etc.) or can be a result of complex reactions of chemicals in the air, such as sulfur dioxide (Werner et. al., 2015). On the other side, the main source of tropospheric ozone (O_3) is represented by hydrocarbons, which are released by plants and soil (Zhang et.al., 2016).

TNO/MACC 2 emissions inventory (the *Netherlands Organisation for Applied Scientific Research/ Monitoring Atmospheric Composition and Climate*), (Visschedijk et al., 2007, Kuenen et al., 2011) contains the internationally verified inventories of emissions, obtained on the basis of country-specific official emissions reported at sectoral level. In air quality modeling, these emissions are divided throughout the year on monthly, weekly and hourly time profiles in order to best reflect real world variations in emission-producing activities. The TNO emission database provides data on the spatial distribution of some important pollutants, such as: NO_x, CO, CH_4, SO_2, NH_3, NMVOC, $PM_{2.5}$ and PM_{10}, for a total of 43 countries and 77 different sources (Figure 1).

The TNOMACC_II emissions inventory is suitable for application in modeling and impact studies related to air pollution policies.

The present paper, aiming at presenting an overall image of pollutant transports over the territory of Romania, is structured as follows: section 2 describes the data and methods used in the numerical forecast of PM_{10} and O_3 over

the Romanian territory, while section 3 outlines the results. Section 4 summarizes the conclusions.

Source: TNO (www.tno.nl)

Figure 1. European countries for which TNO emissions inventory is made

2. Data and methods

The WRF-CHEM air quality forecasting model (version 3.5), comprised of the *Weather Research and Forecasting-WRF* model, coupled with the chemistry module, was integrated into a geographic area centered on Romania, at a spatial resolution of 10 km (Figure 2). The WRF CHEM integration range has 161x161 grid points and the period of time for which the model was applied spans over the months of January and June of 2013 respectively. The model has 35 vertical levels and the running time is 4 hours and 30 minutes for a forecast of 48 hours.

To initiate the WRF-CHEM chemistry module, the PM_{10} values in the TNO emission database[3] have been used and for the start-up of the weather forecasting model, the ECMWF global model outputs have been taken into consideration (Figure 3)[4]. The numerical WRF-CHEM air quality forecasting model uses the following input: TNO emissions database for 2009; exits of the ECMWF global model - 0.125 ° (16 km) horizontal resolution; data of aggregated gravimetric observations for 24 h. As entry data of the model, land surface meteorological parameters (air-temperature-t2m, dew-point temperature-td2m, air-humidity-u10m, wind speed-v10m, mean sea-level pressure-mslp etc.) have been used, as well as meteorological parameters at different pressure levels (from 1000 to 50 hPa),

[3] Nederlandse Organisatie voor toegepast natuurwetenschappelijk onderzoek, www.tno.nl
[4] www.ecmwf.int

relative moisture, temperature, geo-potential height, u and v wind components. For the chemistry module, the initial and boundary conditions have been calculated from emissions extracted from TNO database. The forecasts obtained with the WRF-CHEM model have a 48-hour forecast validity, with one-hour temporary resolution.

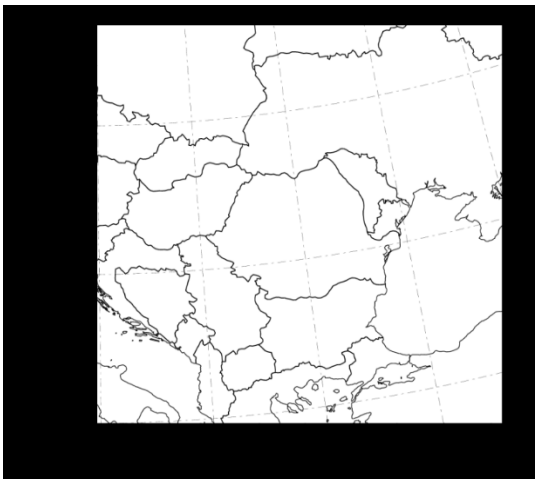


Figure 2. WRF-CHEM model domain

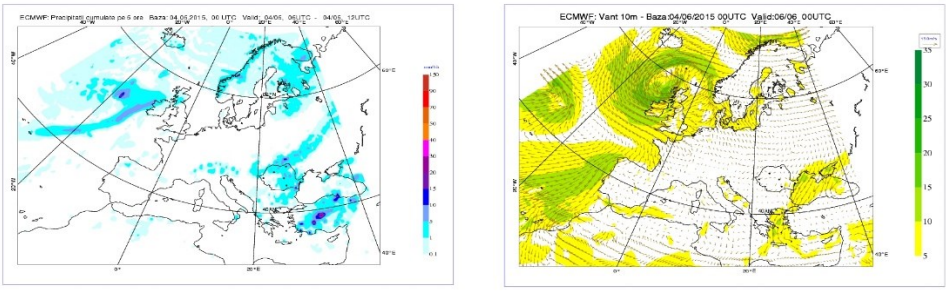


Figure 3. Output data of the ECMWF global model (left) - Cumulative 6-hour precipitation, (right) - Wind at 10 m

The emission processing from the TNO database in the WRF-CHEM model is carried out in two phases; in the first stage, the model transforms the units of the parameters used from tons/year into micrograms/m^3, determining the monthly average values for each day of the week in the regular TNO database grid. In the second phase, the regular grid of the TNO database is interpolated with the WRF-CHEM grid, resulting in monthly average values for each day of the week for any considered parameter. In order to transform the data from the TNO-MACC_II database into the WRF-CHEM model, the EMI2WRF software package provided

by CETEMPS is used to create monthly net CDF files for each pollutant, by performing spatial interpolation and temporal distribution operations (Figure 4).

The pre-processor contains an aggregation module for the emission categories in the model mechanism, as well as a software to create the input files for all months and days considered. In the figure below the input files in the WRF-CHEM model are presented, as extracted from the TNO-MACC_II database, together with the results of the model run.

Emission inventories are relevant tools that can be used to both describe the situation of emissions as well as to decide on the air quality management.

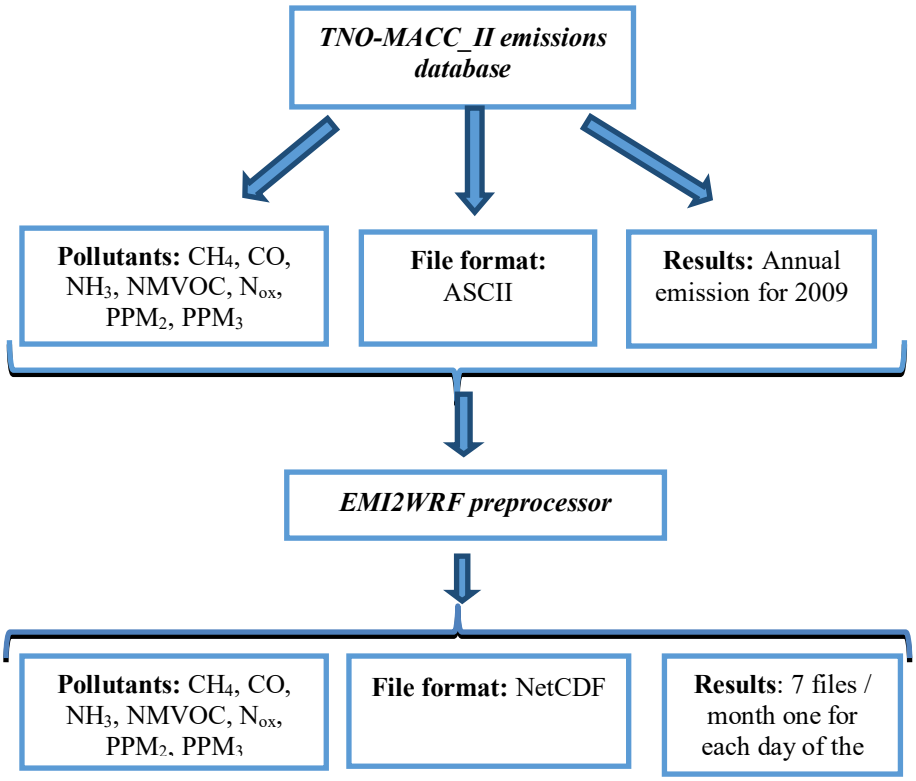
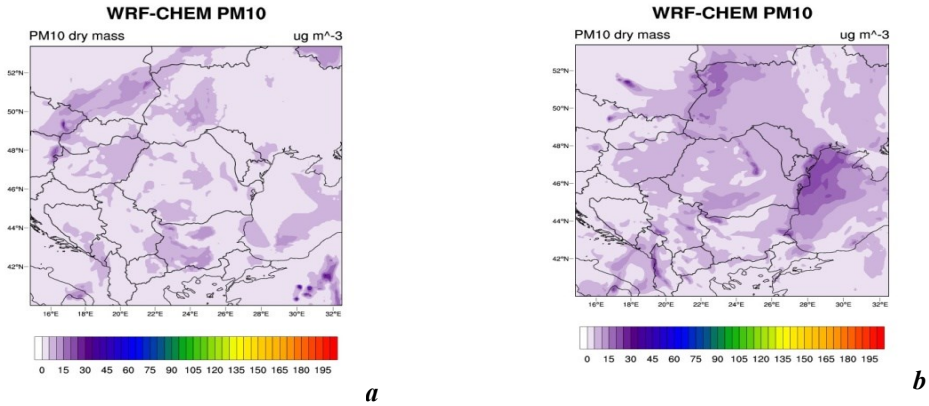


Figure 4. Schematic representation of the use of TNO-MACC_II emission inventory data in the WRF-CHEM

3. Results

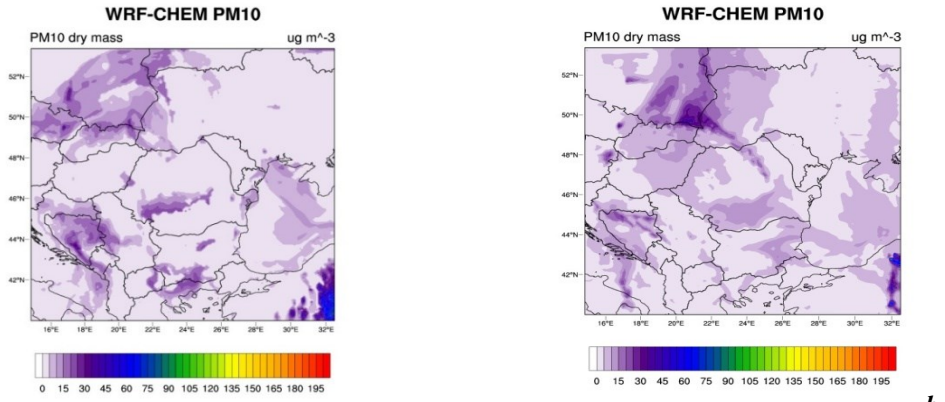
The preliminary results obtained by running the WRF-CHEM model for the Romanian area in the case of **PM_{10}**, for January and June, are presented in the figures below. For the analyzed period, on January 10th, 2013, 09:00 PM (Figure 5-b), the

highest recorded values are ranging from 10 to 25 µg/m^3, while on the same day, at 10:00 AM (Figure 5-a), the modeled values vary between 5 and 10 µg/m^3. It is to be noticed that, at 09:00 PM, the higher values encountered are not concentrated in a single region of the country, although being unevenly distributed, while at 10:00 AM, values not exceeding 15 µg/m^3 are found only in isolated regions. For June 10, 2013, the application of WRF-CHEM Model to simulate PM_{10} concentration at 10:00 AM (Figure 6-b) presents the highest values, comprised between 15 and 20 µg/m^3.



a *b*

Figure 5. PM_{10} forecast for January 10, 2013, resulting from the running of the WRF-CHEM model (a. January 10, 2013, 10:00 AM, b. January 10, 2013, 09:00 PM)



a *b*

Figure 6. The PM_{10} forecast for June 10, 2013, resulting from the running of the WRF-CHEM model (a. June 10, 2013, 09:00 PM, b. June 10, 2013, 10:00 AM)

One can observe that the highest values are concentrated in the southwestern parts of the country, while in the mountain regions, values ranging between 5 to10 µg/m^3 can be identified. The PM_{10} concentration at 09:00 PM (Figure 6-a), reaches

its highest values on a small area located in the center and western parts of the country, while in the rest of the territory there are values that do not exceed 5 $\mu g/m^3$. Compared to the results recorded in winter, during the summer season, a quite reversed situation is evident, with higher values occurring in the morning. Also, with respect to the maximum recorded values, it can be noticed that they vary approximately in the same interval.

In figure 7, there are presented the 24–h average PM_{10} concentrations for two days: 10.01.2013 and 11.01.2013. Both figures show that the highest values are concentrated in the western part of the country and the lowest values are recorded in the mountain area.

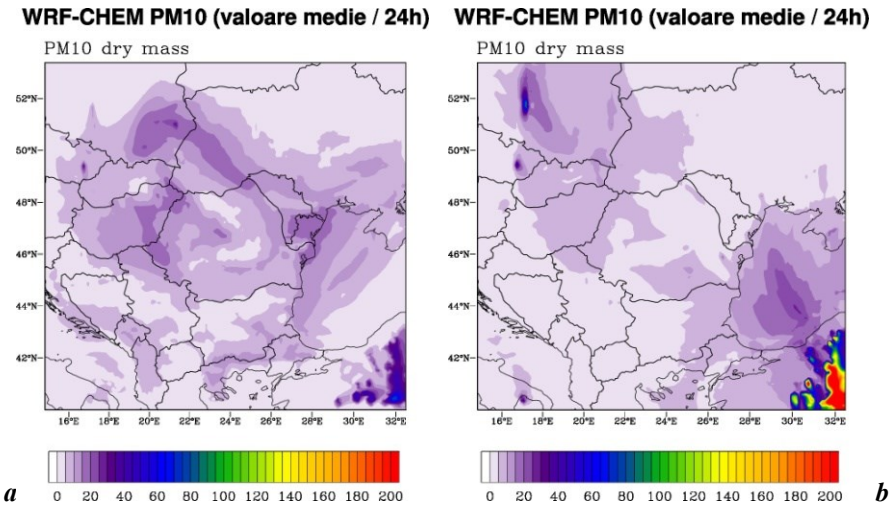


Figure 7. The PM_{10} forecast for January 2013, resulting from the running of the WRF-CHEM model (a. values averaged over 10.01.2013; b. values averaged over 11.01.2013)

In the case of **ground level ozone** (O_3), figures 8 and 9 present the results obtained. It can be noticed that the highest values are recorded in summer, with ordinary values ranging between 60 and 80 mg/m^3, while in winter, on January 10th, the highest values are recorded in the western part of the country, ranging between 20 and 35 mg/m^3.

On 20th January, higher values can be found in most parts of the country, mainly concentrated in the eastern part, with values between 18 and 40 mg/m^3. In the summer season, it can be observed that on 10th June, at 10:00 AM, values as high as 80 mg/m^3 were recorded over most of the country, except for the regions from mountain areas and the north-west.

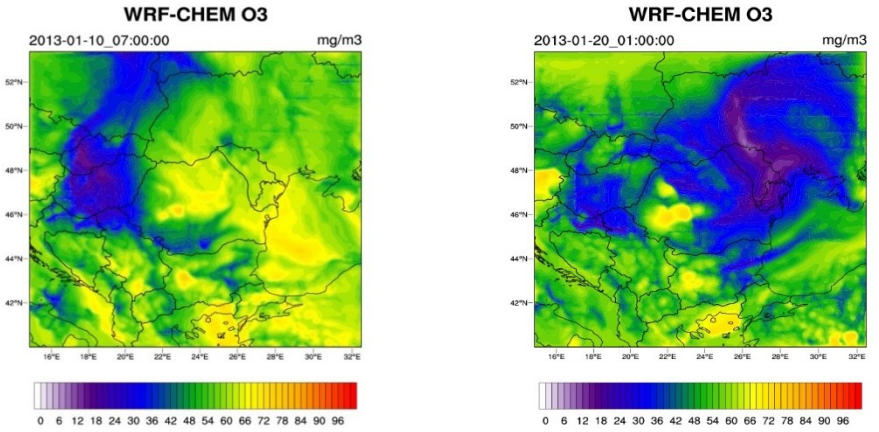


Figure 8. Forecast for O_3 on January 10, 2013 obtained by running the WRF-CHEM model

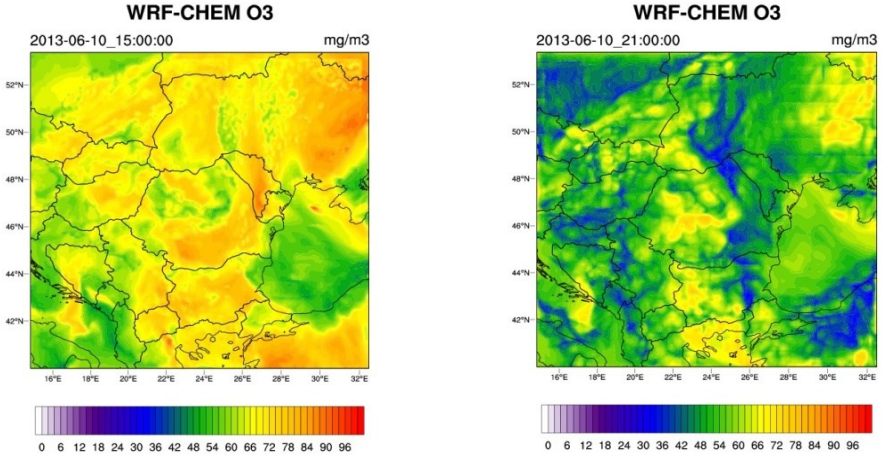


Figure 9. Forecast for O_3 on June 10, 2013 obtained by running the WRF-CHEM model

Conclusions

To highlight the quality of numerical forecasts for PM_{10} and O_3 parameters, they had to be compared with the measurements obtained by gravimetric methods (average daily values of PM_{10}) extracted from the database of the *National Environmental Protection Agency (NEPA)*. In order to make these comparisons, the parameter outputs being representative of the geographical positioning of each NAPM measurement station are extracted from the model outputs.

Numerical air quality forecasting models that can simulate the spatial distribution of reactive gases and particulate matter are already being used in several countries to study the interaction of mineral particles and weather conditions, the dispersion of aerosol biological particles and even accidental pollutant emissions. An advantage of running these types of models within the *National Meteorological Administration* in Romania, is that they can be used to simulate the dispersion and transport of pollutants for two types of emissions: grid emissions and point emissions. The numerical weather forecasting models are important as precursors of air quality models because the latter ones require input data from the former ones, in addition to the grid surface emissions, and also data obtained from meteorological outputs such as: water and dry coat fraction, altitude/pressure (defined by structure of layers), horizontal wind components, temperature, vertical diffusion, water vapor, clouds and precipitation. In the present paper, the air quality model WRF-CHEM at a horizontal resolution of 10 km over the Romanian territory was used for one relevant winter month (January 2013) and one summer month (June 2013). The results show that, during the winter season, the highest PM10 concentrations are ranging between 10 and 25 μg/m3, while in summer, the interval is between15 and 20 $\mu g/m^3$. In the case of ozone-O_3 the simulated values exceed the threshold of 60 mg/m^3, while in winter, the highest values keep well below 20-35 mg/m^3. In order to improve the quality of numerical forecasts that are obtained by using the air quality forecasting models, in recent years a particular emphasis has been put on the implementation and integration of these models with the assimilation of different types of measurements (Žabkar et al., 2015). This study showed only an initial analysis of the performance from the WRF-CHEM model in forecasting PM_{10} and O_3 concentrations for the Romanian territory.

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