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MODELLING OF DRY AND WET DEPOSITION PROCESSES FOR THE SULPHUR AND NITROGEN COMPOUNDS OVER BULGARIA

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***Abstract**: The quality of life is of big importance for human society. Atmospheric composition is one of the most important environmental components, which have a profound influence on human health and quality of life. That is why it is quite natural the surface air pollution concentrations to be most studied. The atmospheric pollution has great impact not only on quality of life, but on the environment as a whole. Therefore, it is important to study the atmospheric compounds dry and wet deposition. The present work is focused on the computer simulations of the dry and wet deposition of the Nx and Sx pollutants. The simulations are performed with the US EPA Models-3 System: Meteorological model WRF; Emission model SMOKE; Atmosphere Composition Model CMAQ for the period 2008 – 2014. The provided model simulations are with horizontal resolution 9 kilometers for the territory of Bulgaria. The NCEP Global Analysis Data meteorological background with 1°x 1°resolution is used as a meteorological background. The models nesting capabilities were applied to downscale the simulations to 9 km resolution. The TNO emission database for year 2005 is used as an input for the Models – 3 System. The model results are compared with the database computed from the EMEP MSC-W model, using a procedure for nudging the results of the 9 km CMAQ simulations, into the EMEP grid with 0.1° x 0.1° resolution.*

Key words: *CMAQ, air quality, WRF, EMEP-MSC-W, EMEP, SMOKE, high-resolution, emission*

INTRODUCTION

The understanding of the air pollution distribution helps us to involve and improve mitigation measures for the human health. The Nitrogen and Sulphur compounds are the ones with adverse health effect on the living beings (Georgieva, 2014). Each of them is a subject on processes of transformation, creation and transportation, which affect their distribution in one or another territory, spatial and temporal scales. There are many observation data acquired by different kind of surface, elevated and satellite measurements. However, to understand their distribution in different spatial and temporal scales, we need to model them with the chemical-transport models. The research community uses different models for studying the air quality in Europe. Two of the most common ones are the CMAQ (Byun and Ching, 1999, Byun and Schere, 2006) and EMEP-MSC-W (Simpson, 2012), run in different temporal and spatial scales, as well as meteorological drivers. The air pollution modelling depends also on the specific emission sources used in the model configurations. The study deals with the air quality features over the territory of Bulgaria. Previous studies on that topic examine the surface concentrations (Chervenkov et al., 2005, 2006, 2008, Chervenkov, 2007) as well as their adverse effect on the human individuals in the country and the surrounding area. Different process determine the concentrations on different height levels - dry deposition, wet deposition, horizontal and vertical advection, horizontal and vertical diffusion, emission, chemical transformation, aerosol processes, and aqueous chemistry. The objective of that research is to study the differences in the high-resolution simulations with the CMAQ and the EMEP-MSC-W models of the Nitrogen (N) and Sulphur (S) dry and wet deposition processes for the territory of Bulgaria over the period 2008 - 2014.

METHODS

The processes accounting to the different contribution of the concentration field changes for each pollutant in the CMAQ model are: horizontal diffusion (HDIF); horizontal advection (HADV); vertical diffusion (VDIF); vertical advection (VADV); dry deposition (DRYDEP); emissions (EMISS); chemical transformations (CHEM); aerosol processes (AERO); cloud processes (CLOUD):

$$\Delta c_i^1 = (\Delta c_i^1)_{hdif} + (\Delta c_i^1)_{vdif} + (\Delta c_i^1)_{hadv} + (\Delta c_i^1)_{vadv} + (\Delta c_i^1)_{drydep} + +(\Delta c_i^1)_{emiss} + (\Delta c_i^1)_{chem} + (\Delta c_i^1)_{cloud} + (\Delta c_i^1)_{aero} \quad (1)$$

The mean concentration change of i^{th} pollutant in for the specific model layer from time t to time $t + \Delta t$ The solution of the transport and transformation equations gives (2)

$$\Delta c_i^1 = \frac{1}{h_1}\int_0^{h_1}\left(c_i(t+\Delta t) - c_i(t)\right)dz \quad (2)$$

The nitrogen depositions contains the contributions from NO_2 (Nitrogen dioxide), NO (Nitrogen oxide), NO_3 (Nitrogen trioxide), N_2O_5 (Dinitrogen pentoxide), HNO_3 (Nitric acid), HONO (Nitrous acid), ANH_4J (Accumulation-mode ammonium mass), ANH_4I (Aitken-mode ammonium mass), ANO_3J (Accumulation-mode nitrate mass), ANO_3I (Aitken-mode aerosol nitrate mass) and NH_3 (Ammonia). The S depositions contains the contribution from SO_2 (Sulphur dioxide), SULF (Sulphate aerosols), ASO_4J (Accumulation-mode aerosol Sulphate mass), and ASO_4I (Aitken-mode aerosol Sulphate mass).

The present work is focused on the computer simulations of the daily dry and wet deposition of the Nx and Sx pollutants with two chemical-transport model systems. The first one is the US EPA Models-3 System. It uses the meteorological model WRF as a driver (Shamarock et al., 2008), the emission preprocessor SMOKE (CEP, 2003, Houyoux and Vukovich, 2009), and MCIP3 as a Meteorology - Chemistry Interface Processor, and the CMAQ as a chemical-transport model. The provided model simulations have hourly time step with horizontal resolution 9 kilometers for the territory of Bulgaria for 2008 – 2014. The NCEP Global Analysis Data meteorological background with 1°x 1°resolution is used as a meteorological background. The models nesting capabilities were applied to downscale the simulations to 9 km resolution. The TNO emission database (Denier 2010) for year 2010 is used as an input for the Models – 3 System for the emission preprocessor. The second model system used for comparison with the previous one is with the Meteorological Synthesizing Centre-West (MSC-W) of the European Monitoring and Evaluation Programme (EMEP). It is a chemical transport model (Simpson et al., 2012), a key tool involving in the European air pollution policy assessments. The model has changed over the years, adding different features, and currently, his horizontal resolution ranging from 5 km to 1 degree with 20 vertical levels. In our study, we use a grid size 0.1° x 0.1°. The EMEP-MSC-W model runs with meteorological fields from the numerical weather prediction system ECMWF-IFS Cycle36r1. The model output is with daily frequency, so we do not need to do further post-processing. For comparison of the models, we use the Normalised Mean Bias noted in the text as NMB (5):

$$NMB = 100 * (\sum_I CMAQ - \sum_I EMEP)/(\sum_I EMEP) \quad (3)$$

RESULTS

The average winter dry nitrogen (N) depositions modelled by the CMAQ are higher and with more heterogeneous spatial structure than by the EMEP (fig. 1). The dry depositions from CMAQ have higher values on the mountain areas. The spatial distribution of the CMAQ total depositions has a structure with characteristics more similar to the dry depositions. The results show that the NMB has a spatial structure follow the topography. The lower terrain parts have lower NMB than the areas with higher elevation. We can see also, that there are three spots of the western part of the domain with a negative NMB. The NMB of the total depositions has similar spatial distribution as for the dry one, but the biases on the mountain and northeastern areas are smaller.

The CMAQ simulates higher average summer N dry as well as wet depositions than the EMEP_MSC_W (fig 2). The dry ones from the CMAQ output are relatively lower in the mountain areas. The wet depositions distribution is opposite. The spatial structure of the EMEP depositions is more homogeneous, as in the winter case. The NMB of the dry and total depositions have not distinctive patterns. The NMB of the wet deposition is negative in the lower terrain forms, and positive in the areas with higher elevation. The values are in the same range as in the winter depositions, except for the wet ones.

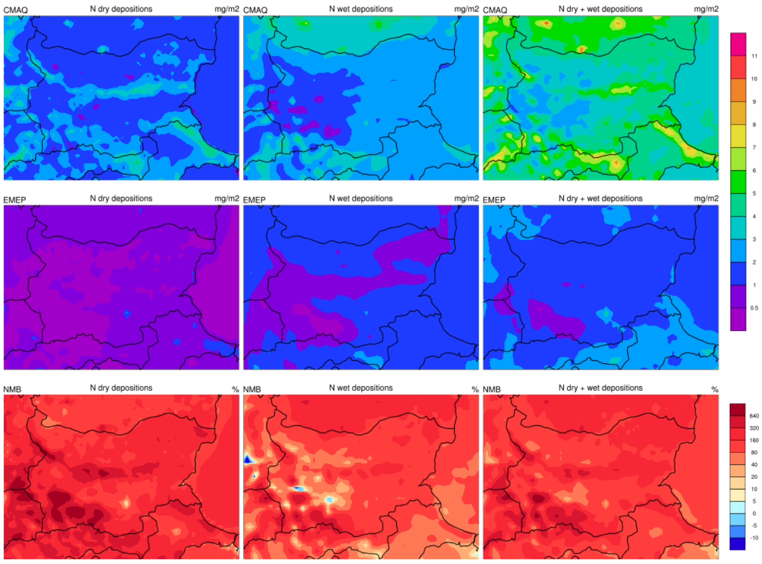

Fig. 1. Average daily dry CMAQ (first column, first row) and EMEP-MSC-W (first column, second row) Nitrogen winter depositions. Average daily wet CMAQ (second column, first row) and EMEP-MSC-W (second column, second row) Nitrogen winter depositions. Average daily total CMAQ (third column, first row) and EMEP-MSC-W (third column, second row) Nitrogen winter depositions. Normalized mean bias (third row) of the dry (first column), wet (second column) and total (third column) depositions.

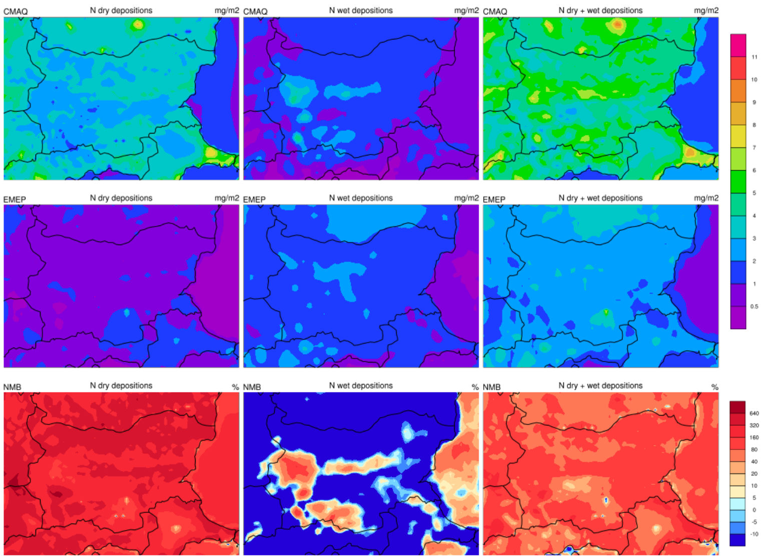

Fig. 2. Average daily dry CMAQ (first column, first row) and EMEP-MSC-W (first column, second row) Nitrogen summer depositions. Average daily wet CMAQ (second column, first row) and EMEP-MSC-W (second column, second row) Nitrogen summer depositions. Average daily total CMAQ (third column, first row) and EMEP-MSC-W (third column, second row) Nitrogen summer depositions. Normalized mean bias (third row) of the dry (first column), wet (second column) and total (third column) depositions.

The spatial distribution of the winter Sulphur (S) depositions is given in the fig. 3. The CMAQ dry S depositions are higher in the mountain and eastern areas, with four spots with values above 9 mg/m^2. The the winter S wet depositions are higher in the southeast. The CMAQ total depositions are distinctively higher in the southeast and the most northern areas. The NMB is mostly positive, with spots of negative values with bigger areas for the wet depositions.

The results for the summer S depositions are given in the fig. 4. The dry ones from the CMAQ are smaller than in the winter, and the EMEP ones do not change so much for that season. The CMAQ wet depositions are higher in the mountain areas. The spots with distinctively higher values are modelled in the western parts of the country by the CMAQ, and in the central by the EMEP-MSC-W. The total emissions repeat and combine these patterns. The NMB does not follow the topography. There are more areas of NMB with negative values, than in the winter, especially for the wet S depositions.

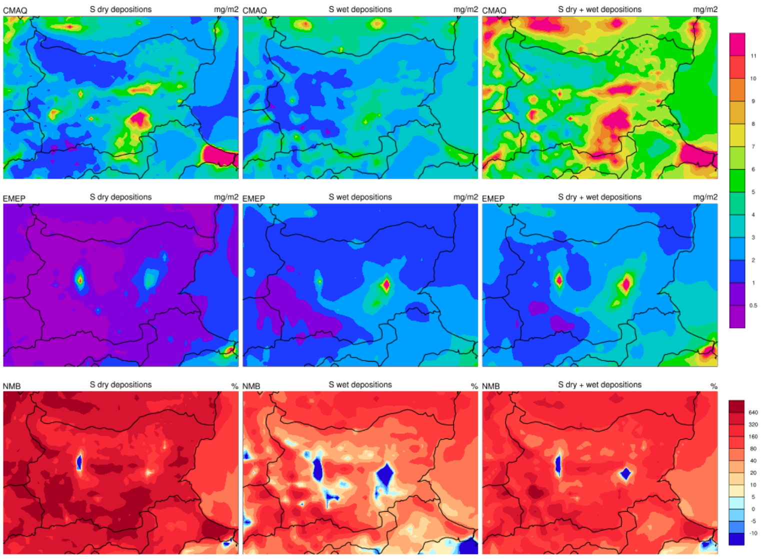

Fig. 3. Average daily dry CMAQ (first column, first row) and EMEP-MSC-W (first column, second row) Sulphur winter depositions. Average daily wet CMAQ (second column, first row) and EMEP-MSC-W (second column, second row) Sulphur winter depositions. Average daily total CMAQ (third column, first row) and EMEP-MSC-W (third column, second row) Sulphur winter depositions. Normalized mean bias (third row) of the dry (first column), wet (second column) and total (third column) depositions.

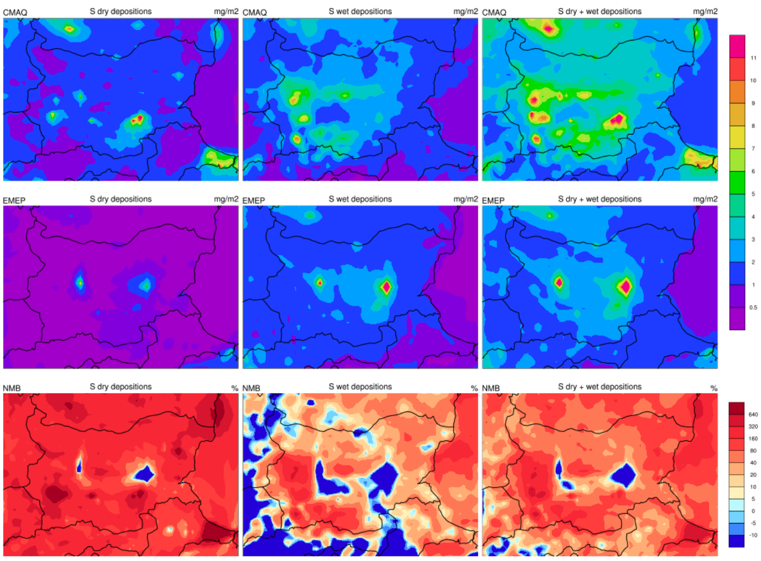

Fig. 4. Average daily dry CMAQ (first column, first row) and EMEP-MSC-W (first column, second row) Sulphur winter depositions. Average daily wet CMAQ (second column, first row) and EMEP-MSC-W (second column, second row) Sulphur winter depositions. Average daily total CMAQ (third column, first row) and EMEP-MSC-W (third column, second row) Sulphur winter depositions. Normalized mean bias (third row) of the dry (first column), wet (second column) and total (third column) depositions.

CONCLUSION

The results show that the output Nitrogen and Sulphur dry depositions of the WRF-MCIP3-CMAQ configuration are higher than the ones from the EMEP-MSC-W model configuration. That statement is valid also for the wet depositions, but to a lesser extent. Three factors determine the simulated differences. The first one is the topography, as we the results shows. The second one is the emission inventory. It implies a difference in the initial conditions for the dry and wet depositions between the two systems. The other main factor is the different meteorology drivers used in the CMAQ and EMEP-MSC-W, which together with the topography features increase or decrease the wet depositions on more or less areas.

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