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CMAQ PERFORMANCE FOR PRECIPITATION CHEMICAL COMPOSITION IN URBAN AND MOUNTAIN SITES IN BULGARIA

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Abstract: This study presents analysis of model performance for chemical composition of precipitation in Sofia region, Bulgaria. The wet deposition simulated by the Bulgarian Chemical Weather Forecast System (BgCwFS) is compared to the experimentally derived wet deposition for the period June – December 2017 at two close but different sites: urban, Sofia city (550 m asl) and mountain, peak Cherni vrah -Vitosha mountain (2230 m asl). Sulphates are the prevailing acidifying species in precipitation samples for both sites. Results for wet deposition of sulphur, oxidised and reduced nitrogen are discussed for some selected synoptic situations analysing air mass HYSPLIT backward trajectories.

Key words: CMAQ, model validation, precipitation chemistry.

INTRODUCTION

Over the last decades precipitation chemistry has been intensively studied worldwide mainly due to concerns over acid deposition, eutrophication, trace metal deposition, damages to forests and vegetation, ecosystem health, and global climate change (Fagerli and Aas, 2008; Torseth et al., 2012; Vet et al., 2014; Pascaud et al, 2016). Atmospheric deposition is part of complex air pollution and environmental processes that link emissions of pollutants, their chemical transformation and sinks, and their effects on the earth's surface. In Bulgaria observations on precipitation chemistry are available only for short term periods and at few locations only (Iordanova, 2010, Hristova, 2017). Numerical simulations using WRF-CMAQ have been carried out recently at the National Institute of Meteorology and Hydrology (NIMH) for wet, dry and total sulphur (S) and nitrogen (N) depositions in Bulgaria for the years 2016 and 2017 (Syrakov, et al., 2019a), applying also the approach of precipitation bias adjustment (PBA) for wet depositions (Syrakov et al., 2019b).

The main objective of this study is to check model performance for sulphur, reduced and oxidized nitrogen wet depositions at two close but different sites: urban, Sofia city (550 m asl) and mountain, peak Cherni vrah (2230 m asl), analysing daily values for the period June – December 2017.

METHODOLOGY

The modelling system

The Bulgarian Chemical Weather Forecasting System (BgCwFS), operationally running at NIMH since 2012, is applied for the simulation of wet, dry and total depositions of different pollutants.

BgCwFS has four main computational modules (Syrakov et al, 2014):

– Meteorological model – the Weather Research and Forecasting Model (WRF v.3.6.1), Skamarock and Klemp (2008);

– Chemical Transport Model - the Community Multi-scale Air Quality model, CMAQ v.4.6, Byun and Schere (2006);

– Interface for linking meteorological data to CTM –MCIP v.3.6

– Emission module – The Sparse Matrix Operator Kernel Emissions Modelling System (SMOKE v.2.4).

The emissions are based on the TNO inventory for 2009 (Kuenen et al. 2014), and Bulgarian national emission inventory for 2010. SMOKE is used partly for calculating biogenic emissions and for merging area sources (AS), large point sources (LPS), and biogenic emission files.

The modelling system is set-up for 5 nested domains: from European scale (81 km grid resolution), down to Sofia city (1 km).

Results for domain d3 ("Bulgaria", 9 km grid resolution) are used in the analysis.

Deposition Calculations

The atmospheric deposition outputs of CMAQ have been archived in separate files containing hourly values for 29 species, gas and aerosol particles. The S wet deposition (S-WD) is estimated as the sum of the depositions of sulphate (SO_4^{2-}), sulphur dioxide (SO_2) and sulfuric acid. The oxidized nitrogen deposition flux N_{oxi}-WD includes nitrate (NO_3), nitrogen oxide (NO), and nitrogen dioxide (NO_2), HNO_3, HONO, N_2O_5, PNO_3. The reduced nitrogen deposition N_{red} –WD is the sum of ammonia (NH_3) and ammonium (NH_4^+).

Precipitation Bias Adjustment (PBA) for Wet Depositions

Previous studies on model evaluation for wet deposition in Bulgaria (Syrakov et al. 2019b, Georgieva, E. et al., 2019) indicated that BgCwFS overestimates the precipitation amounts. The application of PBA (Appel et al., 2011) as post-processing to model wet depositions at single observational sites has shown positive effect on seasonal and annual depositions. The model wet depositions *WDmod* were linearly corrected by the ratio of the observed *PRobs* to estimated precipitation *PRmod*, following:

$$WD_{mod}^{adj} = WD_{mod} \, . (PR_{obs}/PR_{mod}) \qquad (1)$$

This correction is applied where model daily precipitations are higher than the observed ones.

Observational data

The observational data consist in daily precipitation samples collected with a wet only deposition sampler WADOS according to EMEP methodologies (EMEP, 2001) at the Central Meteorological Observatory Sofia (42.655 N, 23.384 E, at 586 m a.s.l.) and with passive bulk sampler at High Mountain Observatory - Cherni vrah (42.616 N 23.266 E at 2230 m a.s.l.). The collected daily samples are further analysed for acidity-pH, conductivity-EC, main anions Cl^-, SO_4^{2-} , NO_3^-, cation NH_4^+ and elements Ca, Mg, K, Na, Fe, Si, Zn.

RESULTS AND DISCUSSION

Chemical analysis of the samples

The mean anion and cation concentrations ($mg.l^{-1}$) and the contribution of each ion to total ionic mass (%) in precipitation samples at the two sites for the whole period June to December 2017 are presented on Fig. 1. The concentration of nss_SO_4^{2-} has been estimated by correction based on the mass concentrations of the sodium in the seawater: *[nss_SO_4^{2-}] = [SO_4^{2-}]-(0.25× [Na]),* according to WMO GAW Methodology, 2004. The mean concentrations of all elements are generally higher for the mountain site than for the urban one. Generally, nss_SO_4^{2-} is found to be the dominant anion in precipitation samples for both sites (39.6% for Sofia and 31.2% for Cherni Vruh), followed by NO_3^- (21.6% and 25.6%) and Cl^-(%).

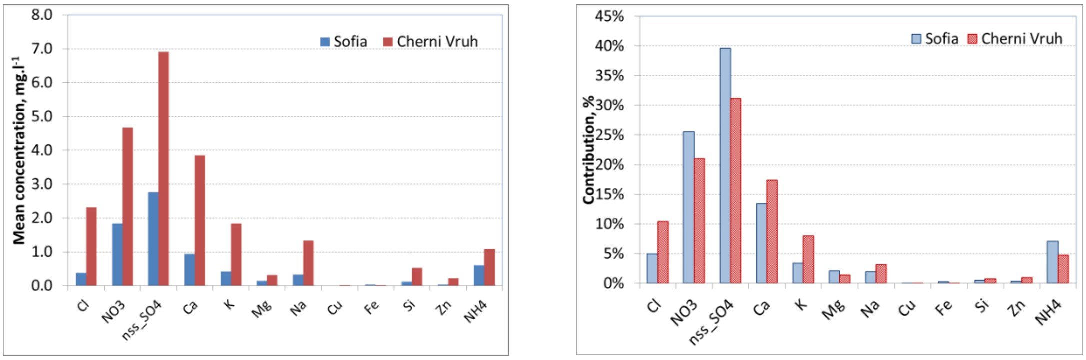

Fig.1. Mean concentrations (right) and percentage contribution (left) for precipitations in the period June – December 2017

The predominant cation is Ca representing 13.4% for Sofia and 17.4% for Cherni Vruh. The contribution of ammonium ions is 7.12% (Sofia) and 4.75% (Cherni Vruh). The orders in percentage contribution of different elements for both sites are: Sofia: $nss\_SO_4^{2-} > NO_3^- > Ca > NH_4^+ > K > Cl > Na > Mg > Si > Fe > Zn > Cu$ and for Cherni Vruh: $nss\_SO_4^{2-} > NO_3^- > Ca > Cl > K > Na > NH_4^+ > Si > Mg > Zn > Fe > Cu$.

Comparison between model and observed depositions

The comparison of two model types of model depositions (without PBA and with PBA, the last noted as "adjusted"), to observed depositions for S_WD, N_{oxi}-WD and N_{red}-WD on monthly basis is presented in Fig. 2, respectively for the two sites. The model simulates correctly the prevalence of S over N depositions for both sites. Higher values for model S, N_{ox} and N_{red} wet depositions at Cherni Vruh compared to those for Sofia is observed.

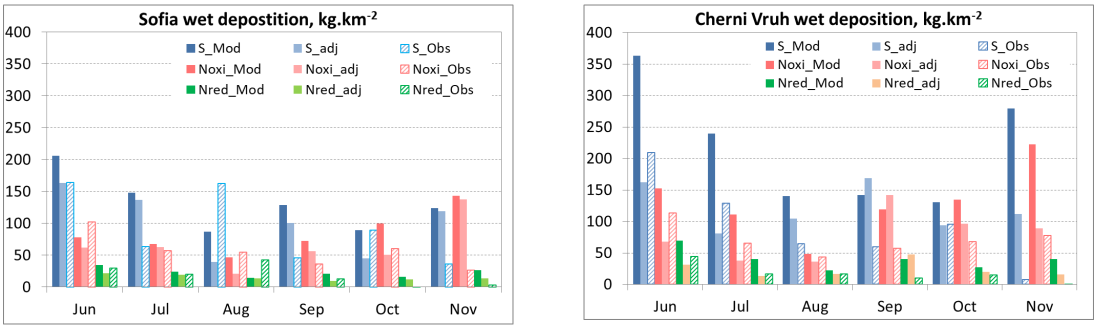

Figure 2. Monthly modelled (_Mod and _adj) and observed (_obs) wet deposition at Sofia (left) and Cherni vruh (right)

The highest normalized mean bias (NMB) for S_WD, N_{oxi}-WD and N_{red}-WD for both sites is obtained for November. The lowest NMB for S_WD is in October, for N_{oxi}-WD - August and for N_{red}-WD - June.

Long range effects for some selected periods

The origin of air masses was examined by means of back-trajectories using the model HYSPLIT (Stein et al, 2015, Rolph et al., 2017) for three selected periods: 16-17 June, 09-13 August and 26-28. September, 2017.

The synoptic situation during the period 16-17.06.2017 was characterised by the passage of a fast cold front from NW (Fig.3a). Simulated and observed daily wet depositions at Sofia have similar values (S_Mod=11.5 kgkm^{-2}, S_Obs=10.07 kgkm^{-2}, N_{ox}_Mod=6.4 kgkm^{-2}, N_{ox}_Obs=7.77 kgkm^{-2}, N_{red}_Mod=2.30 kgkm^{-2}, N_{red}_Obs=3.82 kgkm^{-2}). The situation on 12-13.08.2017 was typical for August, when anticyclonic weather conditions are influenced by low pressure systems passing southward of the country. Due to the atmospheric instability convective storms occur and the precipitation has a patchy character (difficult to be simulated by the model) (Fig.3b). The chemical composition was influenced by forest fires in the western part of the Balkans (unusually high concentration of K was detected in the samples from Sofia. There were number of fires in the period 09-13.08.2017 over SE Europe (see satellite image from VIIRS 375 m, https://firms.modaps.eosdis.nasa.gov/).

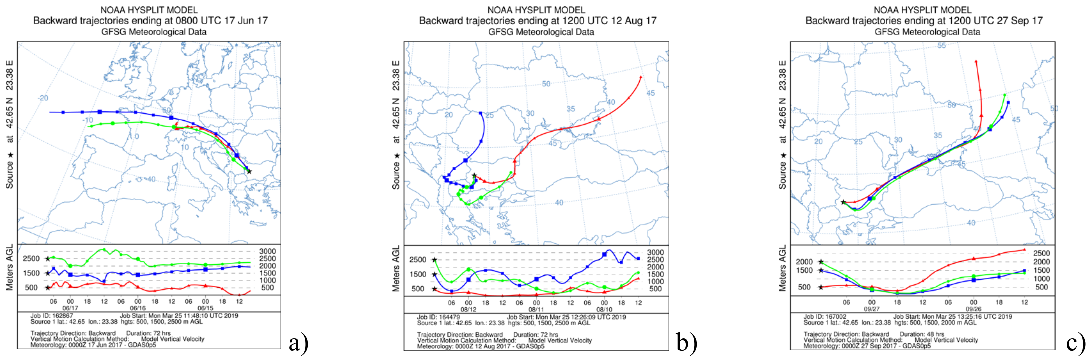

Figure 3 Back-trajectories for periods: a) 16-17.06.2017, b) 12-13.08.2017 and c) 26-28.09.2017

In the period 26-28.09.2017 a large anticyclone was present at surface level, while a low pressure through was situated southward of the Balkans.The flow towrads Sofia and Cherni vruh was from E-SE (Fig.3c). The chemical composition showed eleveated levels of *Cu*, most likely due to the copper smelter plant situated eastward of Sofia. Simulated and observed daily wet depositions at Sofia for 27 September have similar values (S_Mod=25.63 kgkm^{-2}, S_Obs=21.98 kgkm^{-2}, N_{ox}_Mod=16.66 kgkm^{-2}, N_{ox}_Obs=20.50 kgkm^{-2}, N_{red}_Mod=3.79 kgkm^{-2}, N_{red}_Obs=8.15 kgkm^{-2}).

CONCLUSIONS

The BgCwFS has been set up for calculations of deposition fluxes using nested domains from European to country and city level. The simulated wet depositions have been compared to observed depositions from two stations –Sofa (urban) and Cherni vruh (mountain) for the period June – November 2017. Sulphate was prevailing in the collected samples – with share of 39.6% for Sofia and 31.2% for Cherni Vruh, followed by nitrate - 25.6% and 21.6%. The observed S-wet deposition for the study period is estimated as 565 kgkm^{-2} (Sofia) and 566 kgkm^{-2} (Cherni Vruh), while for N_{oxi}-WD is 327.95 kgkm^{-2} (Sofia) and 326.05 kgkm^{-2} (Cherni Vruh), N_{red}-WD is 110.37 kgkm^{-2} (Sofia) and 96.38 kgkm^{-2} (Cherni Vruh). The estimated monthly S wet depositions overestimate the observed ones at both sites - Sofia and Cherni Vruh.

The precipitation bias adjustment method was applied as post processing to simulated monthly wet depositions and showed positive effect. The correction of modelled precipitation values with observations leads to better performance of the model, especially for sulphur depositions.

Results for wet deposition of sulphur, oxidised and reduced nitrogen for three selected synoptic situations have been discussed. The simulated daily S wet deposition slightly overestimates the observed ones for both sites. The values for modelled N wet depositions (N_{oxi} and N_{red}) are lower than those obtained from observations and suggest deficit in model emissions. Further studies are needed for analysing the effects of finer model grid.

In an accompanying paper in this proceeding the results from comparison of EMEP and WRF_CMAQ modelling systems for deposition estimates in Bulgaria for 2016 and 2017 is presented.

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