Modelling of the Seasonal Sulphur and Nitrogen Depositions over the Balkan Peninsula by CMAQ and EMEP-MSC-W

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Abstract The air quality US EPA models-3 system consisting of SMOKE—emission model and pre-processor, MM5—meteorological driver, and CMAQ—chemical-transport model, is used in many studies of the air quality in the Balkan Peninsula, and in particular Bulgaria. It runs in different model resolutions, depending on the domain, from European to city scale. The EMEP-MSC-W model is another chemical transport model, widely used in air quality modelling. Two of the processes involved in the concentration change of some pollutant are the dry and wet depositions. The air quality modelling capability depends on many factors, for example, meteorology and emissions. We study the differences in the simulation of the wet and dry depositions for Nitrogen and Sulphur compounds, between the CMAQ and the EMEP-MSC-W model for a period of 8 years.

Keywords Modelling · CMAQ · EMEP · Pollution · Composition · Air Quality

1 Introduction

The air pollution nowadays forces many countries to take actions for mitigating its adverse effects on human health. Therefore, we need a lot of information, which is increasing in recent years. There are already more direct and indirect data connected to the air quality from different surface-based and satellite-based observing systems. However, we need to understand the different processes involved in the creation, transportation, and transformation of the air pollutant species, which help us to understand their distribution at different spatial and temporal scales. The research community performs these tasks by air quality models systems, with chemical transport models as the main component [4–6]. We use one of these systems with the chemical transport model CMAQ, for modeling the air quality in the Balkan Peninsula. Previous results from air pollution modelling for the Balkan Peninsula and Bulgaria

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are published in a lot of research works [7, 13, 17, 21, 21, 22, 26]. The air composition is formed by several processes, which involve dry deposition, wet deposition, horizontal and vertical advection, horizontal and vertical diffusion, emission, chemical transformation, aerosol processes, and aqueous chemistry [12, 14]. They interact in between and determine the air composition at different scales. There are some studies with CMAQ of the dry and wet deposition and their influence on the precipitation for Bulgaria [15, 16, 24, 25] for different periods up to two years. Another chemical transport model—EMEP-MSC-W is also widely used for air quality studies in Europe [23]. Is the objective of the research is to study the seasonal differences between long-term high-resolution simulations with the CMAQ and the EMEP-MSC-W models of the Nitrogen (N) and Sulphur (S) dry and wet deposition processes in the Balkan Peninsula for a long-term period.

2 Methodology

The study is based on air quality simulations with two chemical transport models over the Balkan Peninsula from 2000 to the 2007 year. One of these simulations is performed with the US EPA Models-3 system, which includes CMAQ (Community Multiscale Air Quality) model [1, 2, 10], SMOKE (Sparse Matrix Operator Kernel Emissions Modelling System) [3, 8, 19] and the regional mesoscale meteorological model MM5. The CMAQ is a numerical chemical transport model for modelling the different processes and their contribution involved in changing the surface and airborne gases and aerosols. That model needs three kinds of input information—initial and boundary conditions, meteorology, and emissions.

We use the regional mesoscale numerical model MM5 for modelling the weather and climate conditions [11, 18] over the Balkan Peninsula. It is a non-hydrostatic high-resolution model, providing the needed raw meteorological output for further processing. We use the nesting capabilities of the MM5, where the output from each outer domain excluding the last one, is used as input for the smaller one. The first and the bigger one (D1) is the European domain with background information, provided from the NCEP Global Analysis Data with $1° \times 1°$ (~81×81 km) horizontal resolution. Our research work is concentrated on the domain D3 with horizontal resolution 9 km and geographically limited to the Balkan Peninsula and some adjacent territories. The output from the MM5 model, need to be reprocessed to the right format for ingesting in the CMAQ. For that purpose, we use the Meteorological—Chemistry Interface Processor—MCIP, which prepares all meteorological input information CMAQ needs.

The emissions from the large source sources and area sources for the whole domain excluding Bulgaria and some adjacent territories are ingested from the TNO high-resolution emission inventory with spatial resolution $0.25° \times 0.125°$ [9] in a longitude-latitude grid, reprocessed from the 50-km grid of the EMEP (European Monitoring and Evaluation Programme) database. The emissions for Bulgaria are from the National Emission Inventory. The CMAQ needs also from biogenic emissions. They are provided from the emission pre-processor SMOKE. The input information is provided from the TNO emissions, the MCIP output, and the land-use database.

The CMAQ model accounts for the following processes with a different contribution to the changing of the concentration field for each pollutant (1): 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). The solution of the transport and transformation equations gives (2) the mean concentration change of ith pollutant in the first model layer from time t to time $t + \Delta t$.

It is presented as a sum of the contribution of the former processes:

$$\begin{aligned}\Delta c_i^1 &= \left(\Delta c_i^1\right)_{\text{hdif}} + \left(\Delta c_i^1\right)_{\text{vdif}} + \left(\Delta c_i^1\right)_{\text{hadv}} + \left(\Delta c_i^1\right)_{\text{vadv}} + \left(\Delta c_i^1\right)_{\text{drydep}} \\ &\quad + \left(\Delta c_i^1\right)_{\text{emiss}} + \left(\Delta c_i^1\right)_{\text{chem}} + \left(\Delta c_i^1\right)_{\text{cloud}} + \left(\Delta c_i^1\right)_{\text{aero}} \end{aligned} \quad (1)$$

$$\Delta c_i^1 = \frac{1}{h_1} \int_0^{h_1} (c_i(t + \Delta t) - c_i(t))\mathrm{d}z \quad (2)$$

We focus on the dry and wet depositions modelled by the CMAQ in this study. The N deposition (3) contains the contribution 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):

$$\begin{aligned}\text{N}_{\text{deposition}} &= \text{NO}_2 + \text{NO} + \text{NO}_3 + \text{N}_2\text{O}_2 + \text{HNO}_3 + \text{HONO} \\ &\quad + \text{ANH}_4\text{J} + \text{ANH}_4\text{I} + \text{ANO}_3\text{J} + \text{ANO}_3\text{I}\end{aligned} \quad (3)$$

The S deposition (4) 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):

$$\text{S}_{\text{deposition}} = \text{SO}_2 + \text{SULF} + \text{ASO}_4\text{J} + \text{ASO}_4\text{I} \quad (4)$$

The CMAQ deposition output is in 1-h frequency. Therefore, we sum up the hourly values of the N and S components for every day of the simulation, finding the daily deposition values.

The second model 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 [23], a key tool involving in the European air pollution policy assessments. In the beginning, the model covers the whole of Europe with a resolution of about 50 km × 50 km, with vertical levels up to the tropopause (100 hPa). 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° × 0.1° (~14 km). 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 the only further post-processing we need is to re-project it to the CMAQ horizontal resolution with 9 km.

For comparison of the models, we use two kind of error characteristics. The first is Normalised Mean Bias noted as NMB (5):

$$\mathrm{NMB} = \frac{\sum_i M - \sum_i E}{\sum_i E}, \tag{5}$$

and the second is the Mean Bias MB (6):

$$\mathrm{MB} = \frac{1}{n}\sum_i M - \frac{1}{n}\sum_i E \tag{6}$$

The notions in these equations are *i*—*i*th value, *M*—the output form CMAQ, *E*—the output from EMEP-MSC-W. The model simulations are run for the period from 2000 to 2007 years. We calculated the dry and wet depositions for summer (June-July–August) and winter (December-January–February). The results are revealed with the multiyear averaged values of the NMB for each grid point and the annual spatial-averaged values of the bias of the CMAQ output.

3 Results

The results are given for the N depositions and for the S depositions, separated in dry component, wet component, and total (dry + wet) component. The winter multiyear average of the S dry, wet, and total depositions (see Fig. 1) reveals the following features. We can clearly note especially from the dry and the total depositions, the missing of some of the S sources in one model, but not in the other. The most noticeable features in the wet depositions are the missing Southern Italy sources in the CMAQ model output. There are also other sources available in the EMEP model, but with smaller intensity in the other one. We can see from the sum of dry and wet deposition shown on the figure, that the TPP Bobov dol, the TPP Pernik, the Sofia city, the town of Devnia, the Bucharest city and the Istanbul city are noticeable in the CMAQ model output, but not in the EMEP-MSC-W output. There are also other sources in Serbia, Bosnia and Herzegovina and Hungary modelled by the CMAQ, but not by the EMEP. On the other hand, Zlatna Panega and Southern Italy sources

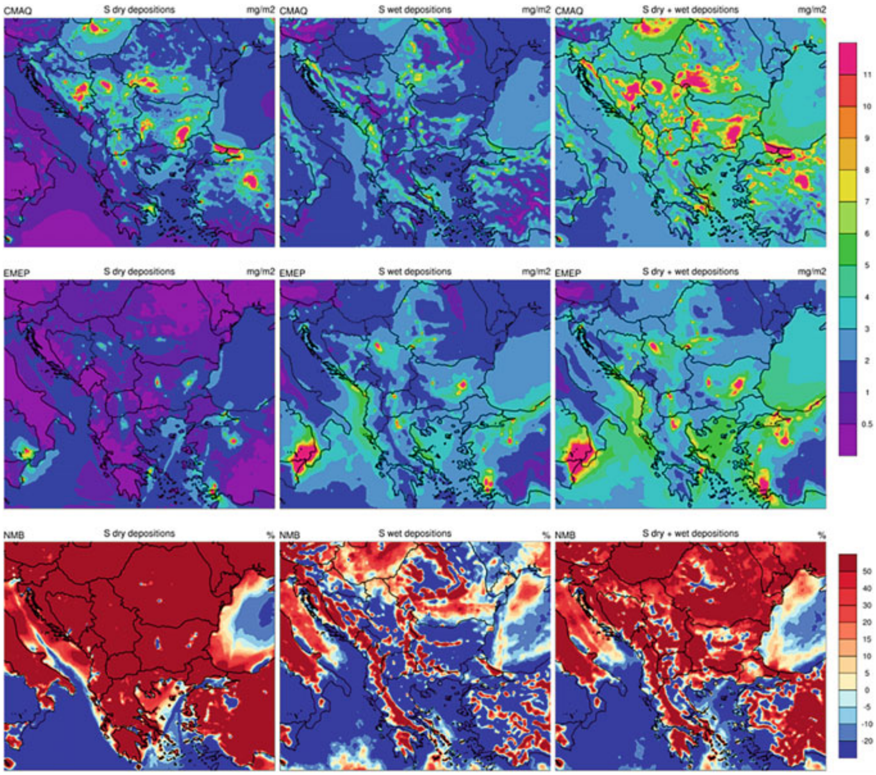

Fig. 1 CMAQ (upper row) multiyear average Sulphur dry deposition (left plot), wet deposition (middle plot) and dry + wet (right plot) in winter. EMEP-MSC-W (middle row) multiyear average Sulphur dry deposition (left plot), wet deposition (middle plot) and dry + wet (right plot) in winter. The dimension of depositions is mg/m^2. Normalized mean bias [%] of the CMAQ model (lowest row) for Sulphur dry deposition (left plot), wet deposition (middle plot) and dry + wet (right plot) in comparison to the EMEP-MSC-W for winter

show up in the EMEP output, but not in the CMAQ one. Thus, the difference between the CMAQ and the EMEP-MSC-W model is due to the difference in the emission inventories, the input meteorological data, and the meteorological driver output. Their influence has a considerable effect on the dry and on the wet deposition modelling capability. The wet deposition in the EMEP-MSC-W model has more intensive local maximums on larger areas around the corresponding sources.

The influence of the meteorological conditions and the orography is notable also in the normalized mean bias field. The normalized mean bias of the dry and total depositions over the land areas reaches 50% and more and only above some of the sources is negative. The normalized mean bias of the wet depositions has more complex structure. The CMAQ and EMEP-MSC-W simulate the annual area-averaged winter dry plus wet Sulphur deposition in a quite similar way from 2001 and 2002, as is shown in Fig. 3 and Table 1. The difference is increasing in the other years. The winter S

Table 1 Seasonal area averaged and multiyear area averaged (YA) Sulphur in winter (DJF) and summer (JJA) (dry + wet) depositions

Year	EMEP dry + wet average S(mg/m^2)		CMAQ dry + wet average S (mg/m^2)		Mean Bias of dry + wet S (mg/m^2)	
	DJF	JJA	DJF	JJA	DJF	JJA
2000	4.423	2.377	3.89	1.401	−0.54	−0.975
2001	3.459	2.829	3.703	2.357	0.245	−0.472
2002	3.778	3.595	3.626	2.07	−0.152	−1.525
2003	4.041	1.859	4.436	1.68	0.395	−0.179
2004	3.536	2.389	4.237	2.207	0.701	−0.182
2005	3.346	2.092	4.337	2.504	0.991	0.412
2006	3.209	2.081	3.794	2.114	0.586	0.033
2007	2.564	1.793	3.828	1.814	1.264	0.021
YA	3.476	2.258	4.047	2.093	0.571	−0.164

bias is increasing from 2000 to 2007, and is positive, which is easy to suggest from the area-averaged total depositions and the CMAQ bias for the whole period.

The results for the summer S depositions (see Fig. 2) have similar patterns, but there are differences. The dry deposition picture show more intensive local sources from the CMAQ model, than the ones in the EMEP. The sources in the Southern Italy are recognized by the EMEP model, but the ones from the Istanbul are spread over a bigger area in the CMAQ output. The summer S wet depositions have smaller intensity than the winter ones. The summer S wet depositions are bigger in the EMEP output. Respectively, they are not so intensive around the sources in CMAQ. The results for the total dry + wet summer S depositions show similar patterns, but the local sources are more intensive than in the two components. The normalized mean bias is generally positive for land dry depositions, except in the proximity of some sources. The results for the wet and total depositions are more similar than in the winter season, with negative bias in the western part, and positive one in the eastern part of the Southeastern Europe. The summer total bias does not follow the orography features, as much as in the winter. Therefore, the main contribution the normalized mean bias is the dry deposition for the winter, and wet deposition for the summer. The area-averaged annual total summer S depositions (see Fig. 3), show negative bias up to 2004 year, and generally non-increasing behavior from 2003 to 2007. These differences could be due to the modelling capability of the circulation features of the EMEP and CMAQ meteorological drivers, the emission inventories, as well as the particular meteorological boundary conditions. The interaction of these factors together with the complex orography of the domain additionally increase these differences. For example, the differences in the contributions of the deposition components in the bias, could be due to the stronger liquid precipitation factor in the summer.

The result for the winter N depositions is shown in Fig. 4. They have different spatial and temporal features from the S ones. As is seen in Fig. 4, the model difference

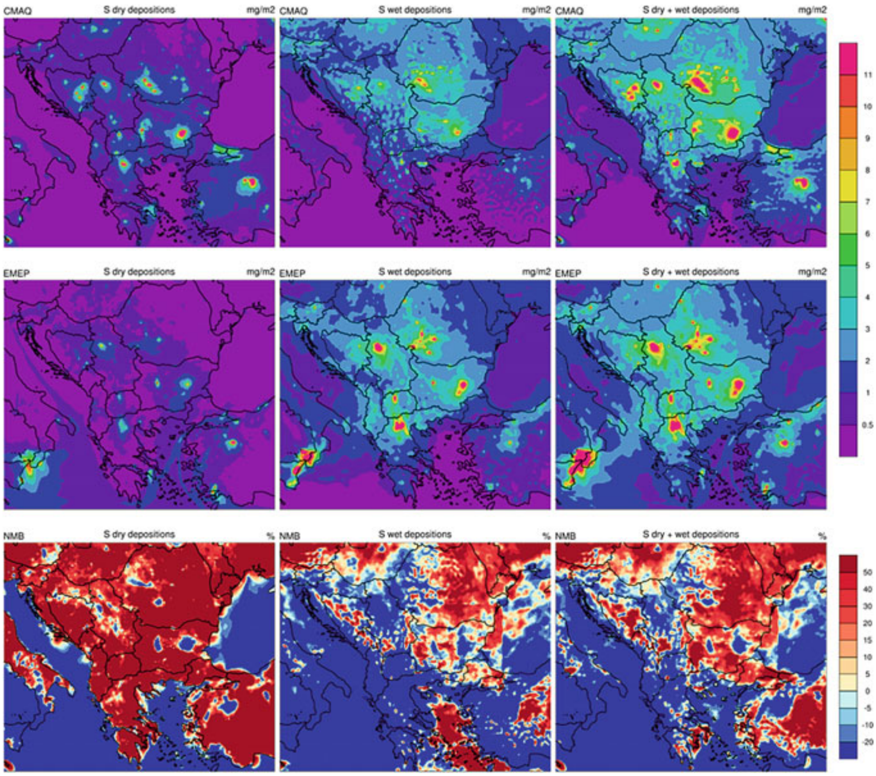

Fig. 2 CMAQ (upper row) multiyear average Sulphur dry deposition (left plot), wet deposition (middle plot) and dry + wet (right plot) in summer. EMEP-MSC-W (middle row) multiyear average Sulphur dry deposition (left plot), wet deposition (middle plot) and dry + wet (right plot) in summer. The dimension of depositions is mg/m^2. Normalized mean bias [%] of the CMAQ model (lowest row) for Sulphur dry deposition (left plot), wet deposition (middle plot) and dry + wet (right plot) in comparison to the EMEP-MSC-W for summer

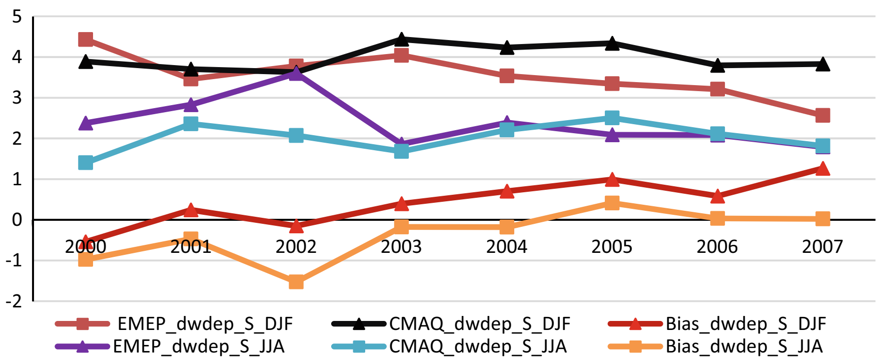

Fig. 3 Seasonal area averaged Sulphur in winter (DJF) and summer (JJA) (dry + wet) depositions and mean bias of the CMAQ model

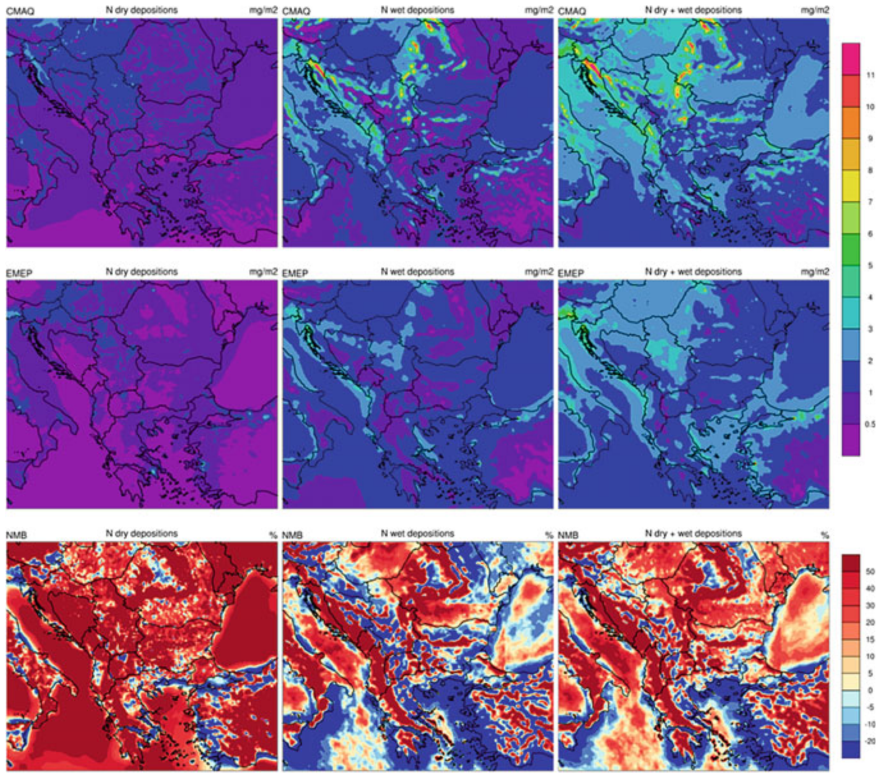

Fig. 4 CMAQ (upper row) multiyear average Nitrogen dry deposition (left plot), wet deposition (middle plot) and dry + wet (right plot) in winter. EMEP-MSC-W (middle row) multiyear average nitrogen dry deposition (left plot), wet deposition (middle plot) and dry + wet (right plot) in winter. The dimension of depositions is mg/m^2. Normalized mean bias [%] of the CMAQ model (lowest row) for nitrogen dry deposition (left plot), wet deposition (middle plot) and dry + wet (right plot) in comparison to the EMEP-MSC-W for winter

between the dry depositions appears in the clear overestimation of the CMAQ with mean normalized bias more than 40%, in contrast to the EMEP-MSC-W. But, there are some spots of smaller differences and bigger local spatial gradient mainly in the places with lower altitude. Generally, the wet N depositions have bigger values. It is more notable in the CMAQ model output in the northwestern quadrant of the domain. The CMAQ also show bigger local gradients, especially in the areas around Adriatic Sea mad Romania. The spatial gradient of the mean EMEP-MSC-W wet deposition is smaller than the CMAQ one. Their normalized mean bias have a substantial local gradients, influenced by the orography. The lower terrain forms except Adriatic Sea are characterized mainly by negative normalized bias, and the higher terrain ones by positive. As in the case of S depositions, the wet ones have bigger influence on the total dry plus wet depositions.

Table 2 Seasonal area averaged and multiyear area averaged (YA) Nitrogen in winter (DJF) and summer (JJA) (dry + wet) depositions

Year	EMEP dry + wet average N (mg/m^2)		CMAQ dry + wet average N (mg/m^2)		Mean Bias of dry + wet N (mg/m^2)	
	DJF	JJA	DJF	JJA	DJF	JJA
2000	1.994	2.074	2.366	1.738	0.372	−0.336
2001	1.785	2.274	2.149	2.37	0.364	0.096
2002	1.82	2.523	2.128	1.828	0.308	−0.695
2003	1.962	2.249	2.282	1.881	0.32	−0.368
2004	1.953	2.516	2.438	2.24	0.484	−0.276
2005	2.202	2.505	2.54	2.321	0.338	−0.18
2006	1.891	2.262	1.992	1.967	0.102	−0.295
2007	1.721	2.102	2.275	1.938	0.555	−0.164
YA	1.954	2.346	2.295	2.081	0.341	−0.265

The results for the normalized mean biases of the dry, wet, and sum of the dry and wet depositions (see Fig. 4) suggest a substantial influence of the meteorological driver, and the orography on the spatial distribution of the mean wet deposition. The normalized mean bias of the mean dry deposition reaches 50% on the land and marine areas, and is negative in few places. The normalized mean bias of the sum of the dry and wet depositions has a similar spatial stricture with the one of the wet deposition, but with more areas with positive one, because of the influence of the dry deposition. The results for the seasonal area averaged total N depositions and the mean bias (see Fig. 6 and Table 2), show little change in years around 0.4 mg/m^2, with a minimum in 2007.

The results for the N summer depositions are depicted in the Fig. 5. The CMAQ overestimate the dry depositions in comparison to the EMEP model. It is notable in the northwestern parts of the continental domain areas. Generally, the CMAQ dry depositions are bigger than the winter ones, which applies to a lesser extent for the EMEP model. The normalized mean bias is positive in the land areas, and negative in the marine ones. There are almost no exceptions.

The wet depositions however, show a different picture. The ones form the EMEP are bigger than the ones modeled by the CMAQ. The normalized mean bias of wet depositions (Fig. 5) is negative in almost all areas. That suggests little influence of orography and emission inventories, and significant one of the meteorological driver, which in the case of EMEP gives bigger precipitations. The two model outputs give more similar total depositions than in the winter case. The northwestern parts are characterized by relatively bigger values in comparison with other areas, which is more notable than the dry and wet depositions. The normalized mean bias is negative in the marine areas, and some land ones. It is positive in the eastern parts of the Balkan Peninsula, in most parts of the Bulgaria and southern Italy. Therefore, we suggest that the model differences in the meteorological drivers are modulated by the differences

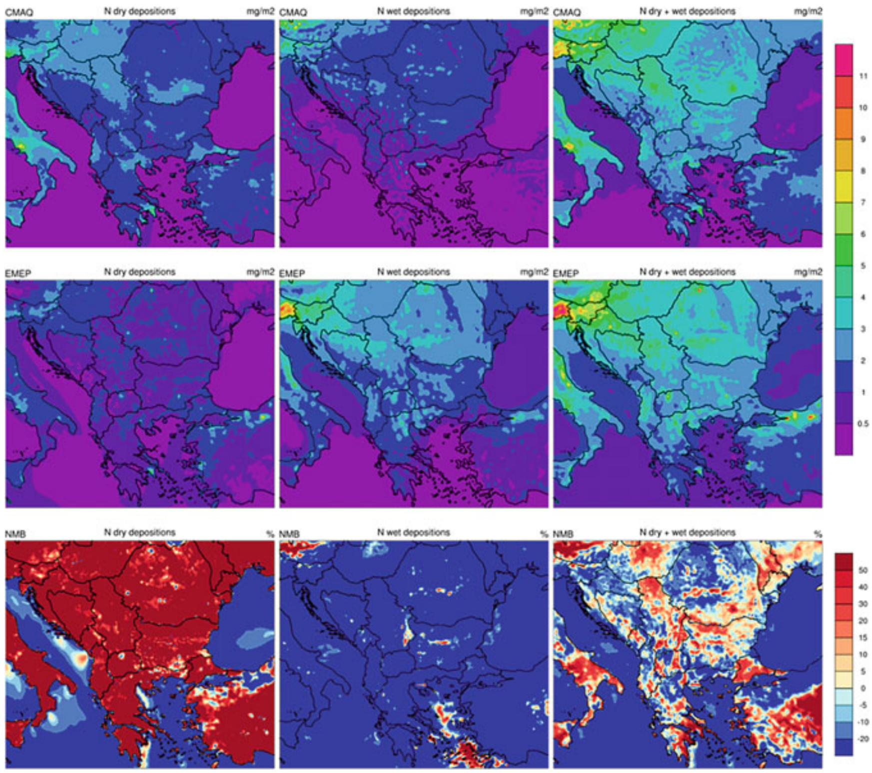

Fig. 5 CMAQ (upper row) multiyear average Nitrogen dry deposition (left plot), wet deposition (middle plot) and dry + wet (right plot) in summer. EMEP-MSC-W (middle row) multiyear average nitrogen dry deposition (left plot), wet deposition (middle plot) and dry + wet (right plot) in summer. The dimension of depositions is mg/m^2. Normalized mean bias [%] of the CMAQ model (lowest row) for nitrogen dry deposition (left plot), wet deposition (middle plot) and dry + wet (right plot) in comparison to the EMEP-MSC-W for summer

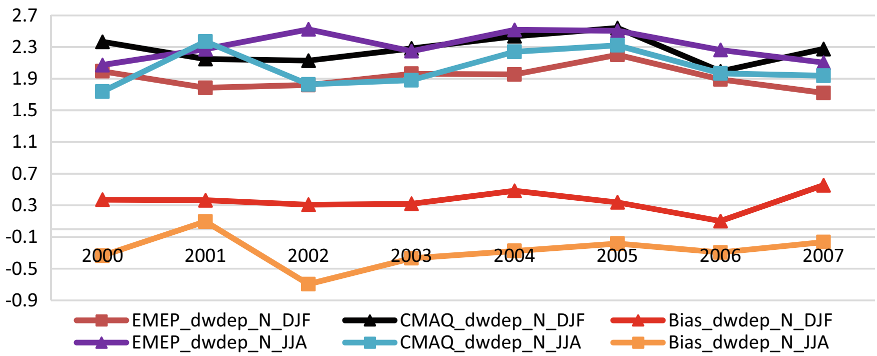

Fig. 6 Seasonal area averaged Nitrogen in winter (DJF) and summer (JJA) (dry + wet) depositions and mean bias of the CMAQ model

between the emissions inventories. The mean bias of the seasonal area averaged summer total depositions (see Fig. 6) is generally negative except in 2001 while winter one is strictly positive. It could be due to the changes in emissions and/or weather features at different scales.

4 Conclusion

The results suggest that the Nitrogen annual area-averaged total depositions are represented more similarly by the two models, than the Sulphur ones. There is a large orography influence on the sum of dry and wet deposition for both groups of chemical species mainly in the winter. Due to the almost equal resolution of both models (EMEP-MSC-W and CMAQ), the orography plays a more important role indirectly through the differences in the model dynamics and physics parameterizations. The emission inventory is the main factor for the biggest differences in the modelled Sulphur depositions and the complex orography is the main factor for the differences in the Nitrogen depositions in the winter. The influence of the meteorological driver and boundary conditions, modulated by the different emission inventories used in both models have the biggest contribution to the normalized mean bias in the summer Nitrogen depositions. The mean bias of the Seasonal averaged total Sulphur depositions increases from 2000 to 2007. The one of the Nitrogen is negative, with smaller absolute values, and with little changes during the years. The variations from 2000 to 2003 are pronounced in both Nitrogen and Sulphur total depositions. The study of the reasons for that models behavior in the pointed period is one of our future plans.

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