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## ANALYSIS OF POSSIBLE CLIMATE CHANGE IMPACTS ON AIR POLLUTION BY SULFUR SPECIES OVER CENTRAL-EASTERN EUROPE

High resolution, complex modelling system built with regional climate model (RegCM3), original emission model (EMIL) and air quality model CAMx was employed for analysing projected climate change impacts on concentrations and depositions of sulfur compounds ( $SO_x$ ) over Central and Eastern Europe. With employment of constant emission rates, results show a slight increase of  $SO_2$  concentrations in the future, as well as increase of  $SO_x$  deposition in the mountains and decrease in central and eastern parts of Poland. Projections indicate also slight changes in a number of days and hours during the calendar year with  $SO_2$  levels exceeding European limit values. The biggest changes are evident in the vicinity of large point emission sources.

## 1. INTRODUCTION

Climate change and air pollution are mutually connected. Since 2007, it is very likely (with probability higher then 90%) that most of the observed increase in globally averaged temperatures since the mid-20th century is due to the observed increase in anthropogenic greenhouse gases (GHGs) concentrations [1]. On the other hand, climate change may affect exposures to air pollutants by many ways [2] such as affecting weather and thereby local and regional pollution concentrations (a), affecting anthropogenic emissions, including adaptive responses involving increased fuel combustion for fossil fuel fired power generation (b), affecting natural sources of air pollutant emissions (c), and changing the distribution and types of airborne allergens (d). Local weather patterns influence atmospheric chemical reactions as well as atmospheric transport and deposition processes. In addition, the chemical composition of the atmosphere may in turn have a feedback effect on the local climate. Weather is also

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associated with energy demands (e.g. for space heating and cooling) that could alter patterns of fossil fuel combustion. In particular, individual responses to extremely hot/extremely cold weather can result in large increases in air conditioner use/fossil fuel use for heating purposes.

According to the latest Intergovernmental Panel of Climate Change (IPCC) Assessment Report [1], warming of the climate system is unequivocal, as is now evident from observations of increases in global average air and ocean temperatures, widespread melting of snow and ice, and rising global mean sea level.

Research in the last years has led to a substantial improvement of our understanding of the physicochemical behaviour of the atmospheric and climate systems, as well as impacts to human health and ecosystems. Moreover, a large amount of information has been collected, and modern tools developed, that may be applied to assess both the air quality at regional and local scales, as well as global and regional climate changes. General circulation models (GCMs), also referred to as global climate models are used to simulate long term changes of climate as a result of slow changes in some boundary conditions or anthropogenically driven changes such as the GHGs concentrations. To simulate climate changes in a statistical sense (i.e., the means and variability), GCM's are run for longer periods (years, decades). Most climate models are large dynamical deterministic systems involving a million variables on huge computers. They can reproduce reasonably well climate features on large scales (global and continental) but their accuracy decreases when proceeding from continental to regional and local scales because of the lack of resolution. However, in many applications, particularly related to the assessment of climate change impacts, the information on surface climate change at regional to local scale is fundamental. For this end, during the last decade, regional climate models (RCM) have been increasingly used to examine climate variations at scales that are not resolved by global models (e.g. [3-5]). To the extent that they produce realistic climate simulations, such models can be powerful tools in the studies of regional climate impacts. RCM are usually working with fine spatial resolution of 10-60 km.

For studying climate change impacts on air quality, the modelling systems coupling GCM/RCM with complex air quality models (AQM) are being used. The most widely AQM used in such coupled systems are, in present, the so-called third generation photochemical Eulerian grid models, namely CAMx (comprehensive air quality model with extensions) and CMAQ (community multiscale air quality) models. The comparison of these models is given in [6].

Most of the studies that have attempted to quantify the potential effects of climate change on air quality examined the impact of increased temperature on global  $O_3$  formation [7, 8]. In general, these studies find that  $O_3$  concentrations increase as temperatures rise, although the estimated magnitude of the effect varies considerably. However only few studies dealed with regional and local scale [9–11].

The aim of this study is to apply coupled RCM-AQM modelling system to model the interaction between climate and air quality by projections of present and future air pollution levels and loads in the target regions on Central-Eastern Europe. Sulphur species having multifaceted impacts on human health, ecosystems and radiation budget of the atmosphere are chosen as key species.

## 2. METHODS

#### 2.1. DESCRIPTION OF THE MODELLING SYSTEM

Modern modelling system, based on regional climate model, RegCM3, the original emission model, EMIL, and air quality model CAMx were applied for assessing air pollution in present and future climates. The system was implemented for high resolution ( $10 \text{ km} \times 10 \text{ km}$ ) modelling domain, centred over Poland (52.00 °N, 19.30 °E) on a grid with  $120 \times 109$  points. The map projection choice was Lambert conformal. The modelling domain, called hereafter WUT (Warsaw University of Technology) domain, is presented in Fig. 1. The implementation of the modelling system was carried out within the 6. European Union Framework Programme CECILIA (Central and Eastern Europe Climate Change Impact and Vulnerability Assessment).



Fig. 1. WUT modelling domain with areas chosen for assessment of climate change impacts on forest ecosystems

CAMx is a photochemical third generation Eulerian grid model developed at ENVIRON International Corporation (Novato, California) [12]. Simulations have been carried out using CAMx v.4.40. The chemistry mechanism invoked was Carbon Bond, version 4 [13], including 117 reactions – 11 of which are photolytic – and up to 67 species (37 state gases, up to 18 state particulates and 12 radicals). The domain vertical profile contained 12 layers of varying thicknesses, extending up to 450 hPa. Boundary conditions for air quality simulations were calculated with 50 km×50 km

resolution for the European domain at BOKU (Universität für Bodenkultur, Vienna, Austria) under CECILIA project [14].

Regional climate model RegCM3 is a three dimensional hydrostatic model that allows to simulate the climate over a given area. The model was developed at ICTP (The Abdus Salam International Centre for Theoretical Physics) in Trieste. The dynamical core is based on the hydrostatic version of the NCAR-PSU Mesoscale Model version 5 (MM5) [15]. In present study, for 10 km x 10 km resolution simulations, the improved version of RegCM3 for high resolution use, RegCM3-Beta [5], was used. The domain's vertical profile contained 18 layers of varying thickness, extending up to 50 hPa.

Not all meteorological fields from the RegCM3-Beta may constitute direct input to CAMx model. Therefore, pre-processor which prepares the required input fields was developed at Charles University in Prague under CECILIA project. In pre-processor *on-line* emissions of biogenic hydrocarbons (isoprene, monoterpenes) were also calculated as a function of meteorological parameters (temperature and radiation) and the available land use categories, following Guenther et al. [16].

An important part of the study was development of a detailed anthropogenic emission and emission parameters database for Poland. For this purpose, an original emission model EMIL (EMIssion modeL) was developed. The created database of large combustion plants (LCP) with a stack height that is equal or above 100 m ( $h \ge 100$  m) for reference year 2000 consists of the following information: name of the LCP, number of stack, geographical coordinates, installed technological units, stack height and diameter, temperature of exhaust gases, velocity of exhaust gases as well as amount of emission of SO<sub>2</sub>, NO<sub>2</sub> and PM<sub>10</sub>. LCP sources were simulated in CAMx model individually. For area, mobile and point sources with the stack height below 100 m (h < 100 m), the model generates emissions for each cell of computational grid (10 km × 10 km). This is based on a detailed emission sources inventory composed for reference year 2000 in 1 km × 1 km resolution, meteorological data and terrain characteristics. Data on population density, sector-specific activity, fuel demands and characteristics, and sectordependent Polish specific emission factors were collected. For temporal distribution of emissions, the EMIL model applies sector-specific monthly, daily and hourly emission factors.

For other countries belonging to the modelling domain anthropoghenic emission database was prepared at BOKU, based on EMEP inventory (see [14] for details). For the Pannonian countries, a detailed, high resolution, emissions inventory was used. All these emissions were treated as area sources in CAMx simulations.

For the purpose of evaluation of the modelling system, simulations for 2000 year were carried out with regional climate model driven by the ERA40 reanalysis [17]. Then, high resolution simulations were completed for three decadal time slices: 1991–2000 (control decade), 2041–2050 (the so-called near future climate, DEK2) and 2091–2100 (the so-called far future climate, DEK3). RegCM3-Beta was driven by the ECHAM5 GCM under forcing from the SRES A1B IPCC greenhouse gas emission sce-

nario [18]. Global climate model simulations with 25 km  $\times$  25 km resolution as boundary conditions for regional climate simulations were performed at ICTP under CECILIA project. To insolate climate change impacts on air quality, the anthropogenic emission were kept constant at the values of year 2000 for all simulated time slices.

## 2.2. SELECTED FOREST ECOSYSTEMS

In order to assess climate change impacts on selected forest ecosystems in Poland, two areas with a high afforestation rate were selected: Karkonosze domain covering Karkonosze National Park, and Góry Stołowe National Park as well as Kampinos domain covering Kampinoski National Park. Both areas are different in terms of geographical localization and climatic conditions. The WUT modelling domain with localization of selected forested areas is presented in Fig. 1.

## 2.3. AIR QUALITY STANDARDS AND THEIR EXCEEDANCES

Based on WHO guidelines, standards for ambient air pollutants were set in the framework of EU legislation. The latest EU Directive 2008/50/EC states the following standards:

- Limit value (LV) means a level fixed based on scientific knowledge, with the aim of avoiding, preventing or reducing harmful effects on human health and/or the environment as a whole, to be attained within a given period and not to be exceeded once attained.
- Critical level (CLev) means a level fixed based on scientific knowledge, above which direct adverse effects may occur on some receptors, such as trees, other plants or natural ecosystems but not on humans.

By subtracting the limit values/critical levels from the existing concentrations, the so called exceedances are obtained, which are direct estimates of population and/or environmental risk. Exceedance of CLev/LV is defined as a non-negative difference between CLev/LV and the concentration (C); Ex(CLev<sub>t</sub>) = max { $C_t$  – CLev<sub>t</sub>, 0}; Ex(LV<sub>t</sub>) = max { $C_t$  – LV<sub>t</sub>, 0}, where t is the averaging time for concentration, t = 1 h, 8 h, 24 h, 1 year. The EU Directive 2008/50/EC sets two legally binding SO<sub>2</sub> limit values for the protection of human health: 125  $\mu$ g/m<sup>3</sup> for daily mean SO<sub>2</sub> levels and 350  $\mu$ g/m<sup>3</sup> for hourly mean SO<sub>2</sub> levels not to be exceeded on more than 3 and 24 times per calendar year, respectively. The SO<sub>2</sub> critical level for the protection of vegetation was set at the level of 20  $\mu$ g/m<sup>3</sup> for both calendar year and winter season (from 1 October to 31 March).

## 3. RESULTS

## 3.1. EVALUATION OF THE MODELLING SYSTEM

Evaluation of the modelling system was carried out for 2000 year with RegCM3-Beta driven by reanalysis ERA40 meteorological fields. Annual mean SO<sub>2</sub> concentra-

tions were evaluated using observations from EMEP (http://www.emep.int) and EIONET-Airbase (http://air-climate.eionet.europa.eu) databases. For evaluation, data from 93 stations, including 5 stations located in Poland, were used. Results indicate a satisfactory performance of the modelling system. The following evaluation measures have been obtained: the correlation coefficient between predicted values and observations r = 0.67, Willmott's index of agreement IA = 0.76, root mean square error RMSE = 2.71  $\mu$ g/m³, normalized mean square error NMSE = 0.45. Finally, FAC2, a fraction of predictions within the factor 2 of observations, was determined as a widely used measure of agreement between model results and observations. For WUT domain, 75% of observation—model pairs lie inside the factor 2 agreement area.

## 3.2. IMPACTS OF CLIMATE CHANGES ON SO<sub>2</sub> LEVELS

Figure 2a shows the distribution of calculated annual mean levels of SO<sub>2</sub> for the control period, while differences between pollutant levels for control and for DEK3 periods are given in Fig. 2b.

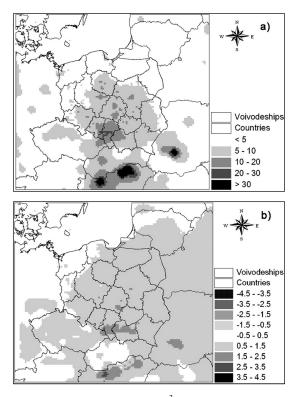


Fig. 2. Annual mean  $SO_2$  concentrations [ $\mu g/m^3$ ] in the control period 1991–2000 (a), and climate change impacts on  $SO_2$  concentrations in terms of the differences [ $\mu g/m^3$ ] for DEK3 (2091–2100) against the control period (b)

For the control period, high  $SO_2$  annual mean levels (up to 30 µg/m³) were obtained for Upper Silesia region in Poland as well as for Hungary and Ukraine, where the levels are the highest and exceed 30 µg/m³. The high  $SO_2$  concentrations calculated for Hungary and Ukraine are due to the treatment of LCP sources as area sources in these countries. In future, higher  $SO_2$  concentrations (up to 3.5 µg/m³) are predicted. The highest increase of  $SO_2$  levels is expected for Upper Silesia region in Poland as well as for Northern Hungary.

# 3.3. IMPACTS OF CLIMATE CHANGES ON EXCEEDANCES OF $\mathrm{SO}_2$ LIMIT VALUES FOR THE PROTECTION OF HUMAN HEALTH

Number of days with daily mean  $SO_2$  levels exceeding 125  $\mu$ g/m³ calculated for the control decade and differences in number of exceedance days between DEK3 and control decade are presented in Fig. 3.

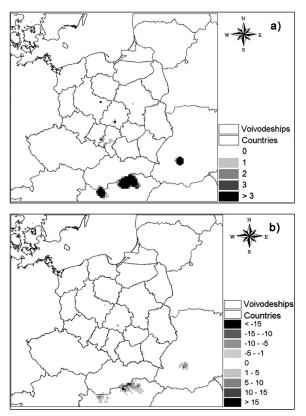


Fig. 3. Number of exceedances [days] over SO<sub>2</sub> daily limit value of 125 μg/m<sup>3</sup> for control decade 1991–2000 (a) and climate change impacts on SO<sub>2</sub> limit value exceedances in terms of the differences [days] for DEK3 (2091–2100) against the control period (b)

On the majority of the WUT domain, no exceedances of the daily LV exist for  $SO_2$  in 1991–2000 period (no days with daily mean concentration exceeding 125  $\mu$ g/m³). The exceedances occur in the surroundings of large combustion plants where the limit value is not met (number of days with daily mean levels over 125  $\mu$ g/m³ is higher than 3). The maximum number of days with average concentration exceeding 125  $\mu$ g/m³ equals to 95 and was calculated for Northern Hungary. For 2091–2100 period there are also no exceedances of the daily LV for  $SO_2$  on the majority of the WUT domain. Only in the surroundings of LCP the number of exceedance days slightly decreases in Poland as well in direct vicinity of LCP and increases in further vicinity of LCP in Hungary and Ukraine. The highest differences were calculated for the vicinity of Mátra Power Plant in Hungary (from -18 to 17 exceedance days).

Number of hours with hourly mean  $SO_2$  levels exceeding 350  $\mu g/m^3$  calculated for the control decade and differences in number of exceedance hours between DEK3 and the control decade are presented in Fig. 4.

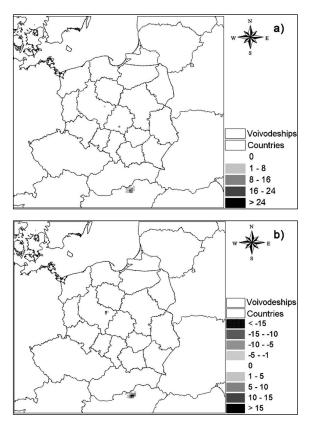


Fig. 4. Number of exceedances [h] over  $SO_2$  hourly limit value of 350  $\mu g/m^3$  for control decade 1991–2000 (a) and climate change impacts on  $SO_2$  limit value exceedances in terms of the differences [h] for DEK3 (2091–2100) against the control period (b)

On the majority of the WUT domain, no exceedances of the hourly LV occur for  $SO_2$  in the control period. The exceedances occur in the surroundings of LCP in Hungary and in Poland, however the number of hours with levels over 350  $\mu g/m^3$  is not higher than 24, thus the LV is met. For 2091–2100 period also no exceedances of the hourly LV for  $SO_2$  occur on the majority of the WUT domain. The climate impact is seen only in few grid points – increase (up to 14 h) in Poland and decrease in the vicinity of Mátra Power Plant in Hungary.

## 3.4. IMPACTS OF CLIMATE CHANGES ON EXCEEDANCES OF $\mathrm{SO}_2$ CRITICAL LEVEL FOR THE PROTECTION OF VEGETATION

Exceedances of  $SO_2$  annual critical levels of 20  $\mu$ g/m<sup>3</sup> calculated in the control period and DEK3 are shown in Figs. 5a, b, respectively.

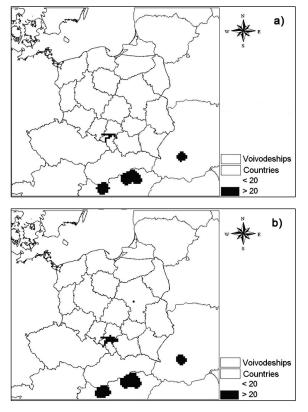


Fig. 5. Exceedances  $[\mu g/m^3]$  of  $SO_2$  annual critical level for the protection of vegetation in the control period 1991–2000 (a) and for DEK3 2091–2100 (b)

In 1991–2000, the annual  $SO_2$  critical level of  $20~\mu g/m^3$  is exceeded only in Upper Silesia industrial region in Poland and for vicinity of LCP in Northern Hungary and Ukraine (maximum value equals to  $51~\mu g/m^3$ ). For far future 2091–2100 period, there is no significant change in the areas and size of exceedances. It can be noticed that exceedances area in Upper Silesia is slightly wider for the end-century decade in comparison to the control period.

## 3.5. IMPACTS OF CLIMATE CHANGES ON SO<sub>x</sub> DEPOSITION OVER SELECTED FOREST ECOSYSTEMS

Deposition of oxidized sulfur ( $SO_x$ ), which is the sum of  $SO_2$  and sulfates,  $SO_4^{2-}$ , was calculated with RegCM3-Beta/EMIL/CAMx modelling system for each grid cell of selected forest areas. Total deposition of sulfur was calculated as the sum of wet and dry deposition. The relevant computational procedures for obtaining a time and space variable dry and wet deposition are included in the CAMx model [12]. Then, trend analysis of the total deposition of  $SO_x$  in the whole 1991–2100 period was performed, based on annual mean deposition calculated for each domain. A linear continuity between analysed periods was assumed.

In a similar way, trend analysis of air temperatures (2 m) and precipitation amounts calculated for 1961–2000, 2021–2050 and 2071–2100 time slices, was conducted (simulations performed by RegCM at CUNI). Concerning precipitation simulation, this is the most difficult task in climate modelling. Nowadays all climate models overestimate precipitation amounts and thus their results cannot be used directly. Consequently, model results are corrected with observations. In the present study, precipitation values were corrected with observations from the Climate Research Unit CRU TS 1.2 [19] database. Figure 6 shows estimated temperature and precipitation values for the whole 1991–2100 period for both selected domains.

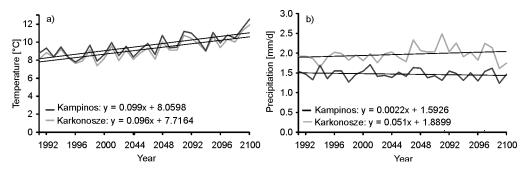


Fig. 6. Projected trend of temperature (a) and precipitation (b) changes for 1991–2100 period for Kampinos and Karkonosze domains

In the 1991–2100 period, increase in temperature is clear and amounts to 2.8 °C for both selected domains. In the same time, precipitation increases by 0.3 mm/day in Karkonosze domain and slightly decreases by 0.05 mm/day in Kampinos domain. Estimated total deposition of oxidized sulfur in the whole 1991–2100 period for both selected domains is presented in Fig. 7.

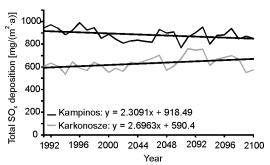


Fig. 7. Projected trend of total deposition of  $SO_x$  [mg S/m<sup>2</sup>] in the 1991–2100 period for Kampinos and Karkonosze domains

For the whole considered period decreasing and increasing trend in total sulfur deposition is projected for Kampinos and Karkonosze domains, respectively. The increase in total sulfur deposition for Karkonosze domain equals to 80 mg S/(m²·year), while the decrease for Kampinos domain equals to 70 mg S/(m²·year).

## 4. DISCUSSION AND CONCLUSIONS

High resolution simulations of climate change impacts on  $SO_2$  concentrations and  $SO_x$  depositions over Central-Eastern Europe were performed. A complex RegCM3-Beta/EMIL/CAMx modelling system was employed for calculations. It turned out to be a useful research tool. The obtained results of  $SO_2$  concentrations in 2000 year reproduce, with satisfactory accuracy, observations from 93 air monitoring stations. Comparison of  $SO_2$  concentrations obtained for the vicinity of LCP sources over Poland and over Ukraine and Hungary indicates better model performance for Poland. This is due to the individual treatment of LCP sources in Poland and employment of LCP parameters for simulation of the subgrid scale rise and spread of the plume. The LCP sources in Hungary and Ukraine were treated as surface area sources. These resulted in significant overestimation of  $SO_2$  concentrations in the vicinity of these sources.

The results of simulations indicate a slight impact of climate change on  $SO_2$  concentrations. Between 2091–2100 and control period a slight increase (up to 2.5–3.5  $\mu$ g/m³) was observed in average  $SO_2$  levels over Central-Eastern Europe. Also exceedances of daily and hourly air quality standards have very similar values and spatial distribution. The biggest changes are visible in the vicinity of LCP sources. In all simulations, the emissions were kept constant and air quality was dependent only on the projected

changes in climate parameters. The results show that SO<sub>2</sub>, as the primary pollutant, slightly subjects to these changes, under assumed A1B IPCC scenario.

In opposite, the impact of climate changes on  $SO_x$  deposition is clearly evident. Trend analysis indicates a decrease in total deposition of sulfur in Kampinos domain equal to 70 mg S/(m²-year) and increase in Karkonosze domain equal to 80 mg S/(m²-year). At the same time, for both domains air temperature increases, while precipitation increases in Karkonosze domain and in Kampinos domain trend line shows no change. The sulfur deposition increase in Kampinos domain is probably due to both an increase in wet deposition (precipitation increase) and in dry deposition. Climate model results (not shown in this study) show increase of average annual wind speed in the future climate for both areas. For Karkonosze domain, it is probably the reason of increase of dry deposition, characteristic of mountains and foothill areas. On the other hand, decrease of sulfur deposition in Kampinos domain is probably associated with the location of the domain in the vicinity of Warsaw agglomeration with large point sources of  $SO_2$ . At higher wind speeds, plume of pollutants from these sources is transported faster over longer distances, resulting in reduction of dry deposition in the analysed domain.

Because many factors affect air quality, the results of this study should not be considered as predictions of future air quality associated with climate change. Rather, they demonstrate the sensitivity of atmospheric air pollutants to changes in specific meteorological variables.

The research presented in this study will be continued. The distribution of  $NO_2$  concentrations and  $NO_x$  deposition, critical loads of sulfur and nutrient nitrogen for selected areas as well as exceedances of critical loads will be examined.

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