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**IMPACT OF DIFFERENT PROCESSES ON THE AIR POLLUTION OVER THE BALKAN
PENINSULA**

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Abstract: Air pollution in cities is a big concern in many countries, although the local authorities take different precautions for mitigation. The meteorological processes in a given area influence the local or regional air composition changes by many mechanisms and are crucial for the air quality in urban areas. The air pollutants' transport and interactions depend on the background weather conditions and their time evolution, as well as the spatial features and types of regional and local emissions. The air quality studies for a specific area can be carried out using different data sources and methods, but the most physically consistent one is by chemical-transport models. They account for different processes determining the air pollutants concentrations by taking into consideration the different sources of emissions, as well as meteorological processes from large to regional and local scales. The studies of the regional air composition are of big importance for decision-makers from the local authorities and other institutions, concerning the air quality. The results from the studies help them to make the right decisions about measures against the worsening of the air quality. The timely reactions of the authorities help to improve the air quality or at least decrease the adverse effects on the general health status of the population in the specific area. They of course depend on the specific country or area, because the air pollutants, their sources, and the processes that change their concentrations vary from one area to another. The countries in the Balkan Peninsula, located in Southeastern Europe have diverse terrain features, and specific air pollution sources, and apply different measures for improving the air quality. The objective of the current research is to study the processes determining the air composition of specific pollutants in the Balkan Peninsula. For that purpose, a modelling system of three models was used: meteorological model - WRF, emission model - SMOKE, and chemical-transport model – CMAQ. The numerical experiments were performed for the period 2008 – 2014.

Key words: *Air Quality, Pollution, Process analysis, CMAQ.*

INTRODUCTION

The meteorological processes in a given area influence the local or regional air composition changes by many mechanisms and are crucial for the air quality in urban areas. The air pollutants' transport and interactions depend on the background weather conditions and their time evolution, as well as the spatial features and types of regional and local emissions. According to ((Gadzhev et al, 2010), (Gadzhev et al, 2015)), the air pollution concentrations of the O₃ and PM_{2.5} over the Balkan Peninsula depend on a small number of processes with complex behavior and contribution, with changing signs in a day. The spatial distribution of O₃, Nitrogen, and Sulphur loads ((Ganev et al., 2008), (Gadzhev et al, 2011)) in Bulgaria, Greece, and Turkey lead to significant transboundary influence appearing in the country-to-country exchange of air pollution. There is also an influence of emission sources on the SO₂, O₃, PM₁₀, and NO₂ concentrations in the Bulgaria-Turkey cross-border area (Syrov et al., 2014). A study of the contribution of the SO_x, NH₃, and other species on the PM_{2.5} formation for the whole of Europe (Clappier et al., 2012) found that SO₂ reduction is efficient in all of the seasons, but the one for the NH₃ depends on the region and season. The countries in the Balkan Peninsula, located in Southeastern Europe have diverse terrain features and specific air pollution sources and apply different measures for improving the air quality. The

objective of the current research is to study the processes determining the air composition of SO₂ and NH₃ in the Balkan Peninsula.

METHODS

The numerical simulations for studying the impact of different processes on air pollution over the Balkan Peninsula were performed. The used system is of three tools for carrying out the modelling of the atmospheric composition. The model WRF version 3.4.1 ((Skamarock et al., 2008), UCAR/NCAR) uses initial and boundary conditions from the 1° x 1° NCEP Global Analysis Data (NCEP FNL, 1999) to produce the input meteorological data for the core of the system – the chemical-transport model CMAQ version 4.6 (CMAS, (Byun et al., 1998), (Byun and Ching, 1999)). The other input type of data for CMAQ are the emissions from the National Emission Inventory for the territory of Bulgaria and for the rest of the Balkan Peninsula domain from the emission for the TNO database (Denier et al., 2010). For their preprocessing is used the model SMOKE (Sparse Matrix Operator Kernel Emissions) (CEP, 2003). The nesting capabilities of the WRF and CMAQ were used, for simulating the air pollution processes by downscaling from D1 domain with 81 km resolution encompassing the European continent, to the D2 domain with a horizontal resolution of 27 km for the Balkan Peninsula. The simulations are carried out for the 2008 – 2014 period. The outputs for the air pollutants SO₂ and NH₃ are with 1-hour frequency.

Sulfur dioxide (SO₂) is part of the sulfur oxides (SO_x) group, formed during the combustion of fuels with high sulfur content. The main anthropogenic source of sulfur dioxide is the combustion of fossil fuels, such as in thermal power plants and household sources (domestic heating). Metallurgy and the chemical industry are also sources of sulfur dioxide pollution. SO₂ and NO_x are key components of "acid rain." Sulfur dioxide enters the body through the respiratory system. The health effects of SO₂ manifest as respiratory disturbances, respiratory diseases, and impairment of the immune defense of the lungs, aggravation of existing respiratory and cardiovascular diseases. Ammonia (NH₃) is a colorless gas with a pungent odor. The main sources of NH₃ are the chemical industry, refrigeration, plants, and agriculture. It is a substance with great practical application. That is why obtaining it cheaply is an important economic and technological task. The effect of NH₃ on human health is expressed in inflammation of the skin, eyes, nose, throat, and lungs. Flue gas desulfurization processes aim at the removal of already-formed sulfur oxides. Modern flue gas treatment technologies are based on sulfur removal through wet, dry, or semi-dry and catalytic chemical processes and are used in different sectors. For example, during desulfurization of ammonia with sulfur oxides, the ammonium sulfate is released by the chemical reaction: 2NH₃ + H₂SO₄ → (NH₄)₂SO₄.

The objective of the study calls for applying the “Integrated process rate analysis” (Gadzhev et al., 2014) for the outputs from the CMAQ. The procedure involves a calculation of the contributions of the horizontal advection (HADV), vertical advection (VADV), horizontal diffusion (HDIF), vertical diffusion (VDIF), dry deposition (DDEP), chemical transformation (CHEM), aerosol processes (AERO), cloud processes (CLDS), and emissions (EMIS) to the change surface pollutant concentration. It is performed by calculation of the change of the mean concentration Δc in the first CMAQ-model layer for every step “Δt”, and for the NH₃ or SO₂ admixtures according to:

$$\Delta c_{tot}^{SO_2} = \Delta c_{HADV}^{SO_2} + \Delta c_{VADV}^{SO_2} + \Delta c_{HDIF}^{SO_2} + \Delta c_{VDIF}^{SO_2} + \Delta c_{DDEP}^{SO_2} + \Delta c_{CHEM}^{SO_2} + \Delta c_{CLOUD}^{SO_2} + \Delta c_{EMIS}^{SO_2} \quad (1)$$

$$\Delta c_{tot}^{NH_3} = \Delta c_{HADV}^{NH_3} + \Delta c_{VADV}^{NH_3} + \Delta c_{HDIF}^{NH_3} + \Delta c_{VDIF}^{NH_3} + \Delta c_{DDEP}^{NH_3} + \Delta c_{AERO}^{NH_3} + \Delta c_{CLOUD}^{NH_3} + \Delta c_{EMIS}^{NH_3} \quad (2)$$

Each member in the right side of equation (1) for the SO₂ and equation (2) for the NH₃, is the change of the mean concentration due to each of the aforementioned processes.

RESULTS

The process analysis of the SO₂ (figure 1) shows the following features. Although the HADV and VADV are not on the same scales, the vertical advection is positive at the places, where the horizontal advection has the largest negative contribution (the sources). The maximum negative contributions of horizontal advection and the positive contribution of vertical advection stand out above the sources (in Bulgaria, these are thermal power plants (TPPs)). The HDIF contribution is almost everywhere negative and tends to zero,

with high sources standing out with maximum negative contribution, while their surroundings are with increased positive contribution and in places - maximum. The VDIF contribution is positive, as it is a maximum above high SO₂ sources. The DDEP contribution is entirely negative, with a maximum above some of the high sources. The contribution of CLDS is small and also entirely negative. The negative contribution of cloud processes/water chemistry is maximum over higher sources and stands out over extensive areas around them. CHEM during most of the day has an almost zero contribution, with only TPPs standing out with a slightly larger negative contribution. The emissions (EMIS) contribution tends to zero over most of the domain, except for some of the largest cities and agglomerations in the region.

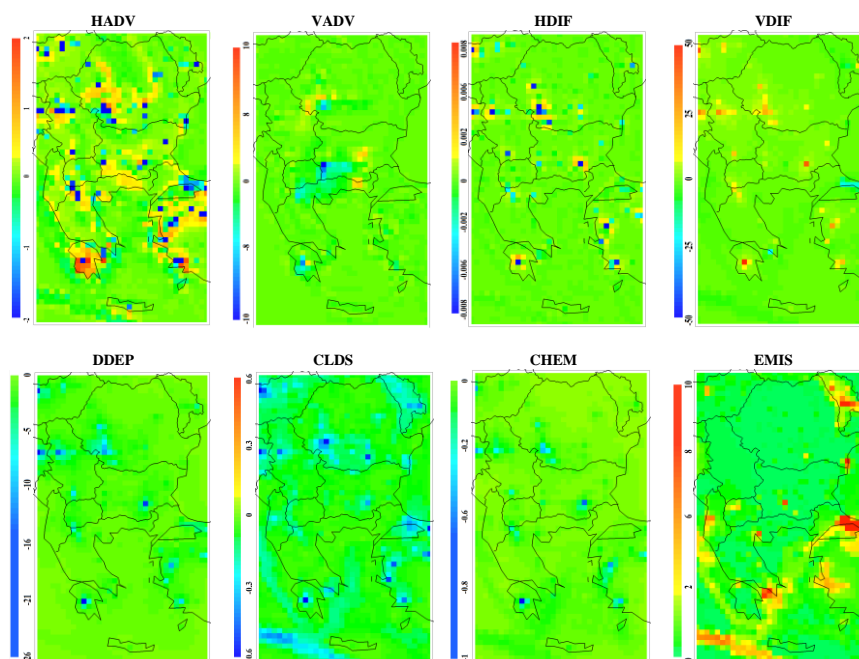


Figure 1. Annual contribution (in $\mu\text{g.m}^{-3}$) of the horizontal advection (HADV), vertical advection (VADV), horizontal diffusion (HDIF), vertical diffusion (VDIF), dry deposition (DDEP), cloud processes (CLDS), chemical transformations (CHEM) and emissions (EMIS) to the formation of the surface SO₂.

The results from the process analysis of the NH₃ (figure 2) suggest that, as for the SO₂, the HADV and VADV are in anti-phase, and in the places where the horizontal advection has the largest negative contribution, the vertical advection is positive. The contribution of horizontal diffusion HDIF is very small and tends to zero, with enterprises standing out with a maximum negative contribution and the surrounding areas with an increased positive contribution. The contribution of VDIF is everywhere completely negative and tends to be zero. It can be seen from the DDEP and CLDS plots the Figure 2, that the contributions of dry deposition and cloud processes/aqueous chemistry are most negative over the plants and agriculture/ livestock facilities, while over the rest they tend to zero almost everywhere. The emissions (EMIS) have a pronounced positive contribution that is maximum over the large regions in the east and north parts of the domain. Above the thermal power plants, the contribution is not only maximum but also observed throughout the day, and this is due to the desulfurization processes. AERO aerosol processes have a completely negative contribution over the entire considered domain.

CONCLUSIONS

The formation of air pollution patterns arises from intricate interactions among various dynamic and chemical processes. Understanding the individual contributions of these processes under diverse meteorological conditions, emission configurations, and temporal profiles is crucial for deciphering atmospheric composition and behavior of air pollutants. Applying the CMAQ "Integrated Process Rate Analysis" option was investigated the role of different dynamic and chemical processes in constructing air

pollution over the Balkans. Through this methodology, was determined the concentration changes of each compound, attributing them to specific processes influencing air pollution levels.

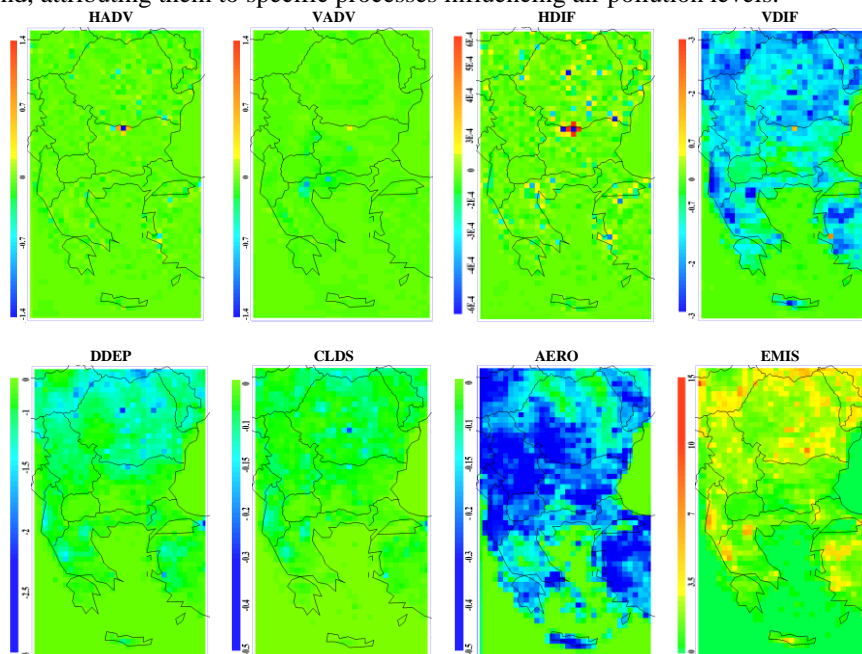


Figure 2. Annual contribution (in $\mu\text{g}\cdot\text{m}^{-3}$) of the horizontal advection (HADV), vertical advection (VADV), horizontal diffusion (HDIF), vertical diffusion (VDIF), dry deposition (DDEP), cloud processes (CLDS), aerosol processes (AERO) and emissions (EMIS) to the formation of the surface NH_3 .

The results from the numerical simulations unveil the complex interaction among these processes, revealing dominant factors with substantial impacts, often with opposing signs and phases. Notably, the magnitude and sign of these contributions vary for different pollutants. While some process contributions are with obvious signs, others demonstrate variability influenced by emission types, weather dynamics, and topographical features.

Some processes as the horizontal and vertical advection, as well as the horizontal diffusion, have either positive or negative contributions, but the vertical diffusion leads to no change or increase of the SO_2 concentration over the region. The cloud processes and dry deposition influence are negative, and the chemical transformations stand out only above the TPPs. The picture for the NH_3 is different mostly with the bigger areas of negative contribution of the vertical diffusion and the distribution of the dry deposition. The horizontal advection and diffusion, as well as the vertical advection contributions, are more spatially homogeneous.

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