

## Introduction

The main scientific challenge of local to regional atmospheric composition pattern modelling probably is the accounting for the strong dependence of concentrations on fluctuations of local and regional meteorological conditions, the complex interaction of transport scales (different life times of the pollutants make it even more complex), uncertainties and responses to emission forcing and boundary conditions, both introducing information noise.

The air pollution transport is subject to different scale phenomena, each characterized by specific atmospheric dynamics mechanisms, chemical transformations, typical time scales etc. The specifics of each transport scale define a set of requirements for appropriate treatment of the pollutants transport and transformation processes, respectively for suitable modelling tools, data bases, scenarios and time scales for air pollution evaluation. The air pollution pattern is formed as a result of interaction of different processes, so knowing the contribution of each for different meteorological conditions and given emission spatial configuration and temporal behaviour. The present study attempts to make some evaluations of the contribution of different processes to the local to regional pollution over the Balkans.

## METHODOLOGY – models, data, domains & nesting

### Modelling tools

The system consists of three components: **MM5** - the 5th generation PSU/NCAR Meso-meteorological Model MM5 used as meteorological pre-processor; **CMAQ** - the Community Multiscale Air Quality System CMAQ; **SMOKE** - the Sparse Matrix Operator Kernel Emissions Modelling System.

### Input data

The large scale meteorological used by the application is the NCEP Global Analysis Data with 1°x1° resolution. At the moment the created database contains all the necessary information since year 2000.

The TNO high resolution inventory are used. The inventory is produced by proper disaggregation of the EMEP 50-km inventory data base. The TNO inventory resolution is 0.125°x0.0625° longitude-latitude, that is on average about 14x7 km. GIS technology is applied as to produce area and large point source input from this data base.

### Domains and nesting

As far as the background meteorological data is the NCEP Global Analysis Data with 1°x1° resolution, it is necessary to use MM5 and CMAQ nesting capabilities as to downscale to 3 km step for the innermost domain. The MM5 pre-processing program TERRAIN was used to define three domains with 81 (D1), 27 (D2) and 9 (D3) km horizontal resolution. These four nested domains were chosen in such a way that the domain with a horizontal resolution of 9 km contains the whole Balkan Peninsula.

### Integrated process rate analysis

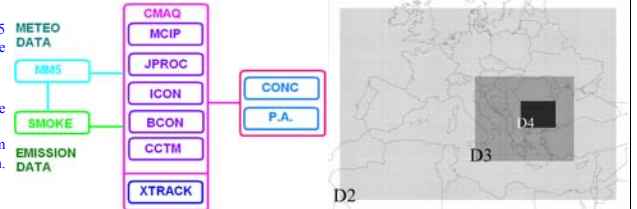
The Models-3 "Integrated Process Rate Analysis" option is applied to discriminate the role of different dynamic and chemical processes for the air pollution pattern formation. The procedure allows the concentration change for each compound for an hour  $\Delta C$  to be presented as a sum of the contribution of the processes, which determine the concentration:

The processes that are considered are: advection, diffusion, mass adjustment, emissions, dry deposition, chemistry, aerosol processes and cloud processes/aqueous chemistry. (1)

$$\Delta C = \sum_{i=1}^N \Delta C_i$$

The Models-3 "Integrated Process Rate Analysis" option is applied to discriminate the role of different dynamic and chemical processes for the pollution from road

and ship transport. The processes that are considered are: advection, diffusion, mass adjustment, emissions, dry deposition, chemistry, aerosol processes and cloud processes/aqueous chemistry.



## SOME EXAMPLES OF PROCESS ANALYSIS SIMULATIONS

The modelling infrastructure has been well validated which allows applying it for air pollution studies for the Balkan region with some trust in the obtained results.

Following the methodology described above, MM5 and CMAQ simulations were carried out for the years 2003- 2009 and the respective process contributions for each day for all the period were calculated. Averaging the process contribution fields over the whole ensemble of results for the respective month produces a diurnal behaviour of given pollution characteristic, which can be interpreted as "typical" for the month (respectively season). The characteristic, which will be demonstrated and discussed as an example further in this paper are the surface process contributions  $\Delta C_i$  and the resulting hourly concentration changes  $\Delta C$  for months January and July.

Plots of the horizontal fields of some of the processes which determine the concentration change of O<sub>3</sub> and PM<sub>2.5</sub> for January and July are shown in Figs. 1, 2. The pattern is indeed very complex. Some typical effects can be followed however. For example the roads, big cities and agglomerations appear as big sinks in the O<sub>3</sub> chemical transformation plot and as big sources in the PM<sub>2.5</sub> aerosol processes plot.

Plots of this kind are rather spectacular and can give a good qualitative impression of the spatial complexity of the different processes contribution. In order to demonstrate the process contribution behaviour and the process interaction in a more simple and easy to comprehend way, the respective fields can be averaged over some domain (in this case the territory of Bulgaria), which makes it possible to jointly follow and compare the diurnal behaviour of the different processes contribution and their resultant hourly surface concentration  $\Delta C$ .

Such plots for NO<sub>x</sub>, SO<sub>2</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> are given in Figures 2-3 for January and July respectively. A detailed description even of these much simpler images will take a lot of space and probably is not necessary. Some more general features could be mentioned, however:

- The hourly surface concentration  $\Delta C$  is determined mainly by a small number of most important the processes (which could be different for different compounds), while the role of the others is minor;
- The temporal behaviour of the processes is also complex;
- For some processes the contribution sign is obvious (like emissions or dry deposition), but some can change their sign during the day;
- For all of the compounds some of the advection/diffusion process have a major role.

## CONCLUSIONS

The numerical experiments performed produced a huge volume of information, which have to be carefully analyzed and generalized so that some final conclusions could be made. The conclusions that can be made at this stage of the studies are that the processes interactions are indeed very complex.

The results produced by the CMAQ "Integrated Process Rate Analysis" demonstrate the very complex behaviour and interaction of the different processes – process contributions change very quickly with time and these changes for the different points on the plane hardly correlate at all. The analysis of the behaviour of different processes does not give simple answer of the question how the air pollution in a given point or region is formed.

The "Integrated Process Rate Analysis" is a fruitful approach, however, so an attempt should be made the evaluation and analysis of the processes to be presented in a more general way. The characteristics demonstrated above are averaged over a large ensemble of simulations. A more extensive statistical treatment – calculating not only the mean process contributions fields, but also standard deviations, skewness, etc. with their dominant temporal modes (seasonal and/or diurnal variations).

The "Integrated Process Rate Analysis" could be also applied in combination with emission sensitivity tests which can outline the impact of different emission categories on the processes by which the air pollution is formed.

Studying the impact of the numerical special resolution to "Integrated Process Rate Analysis" results seems to be a promising task as well.

## ACKNOWLEDGEMENTS:

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## HARMO13

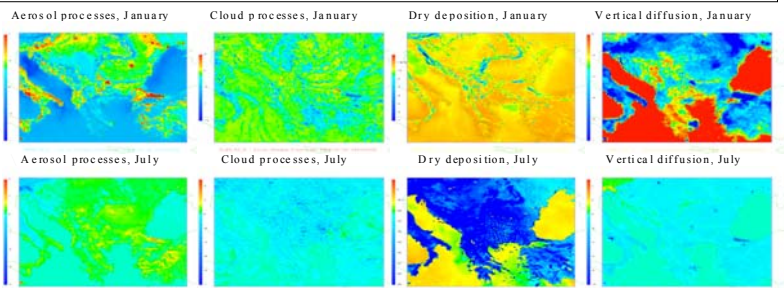


Figure 1. Plots of the contribution [µg/m<sup>3</sup>] of Aerosol, Cloud, dry deposition and Vertical diffusion processes to the formation of PM<sub>2.5</sub> at 16h of a "typical" day in January and July.

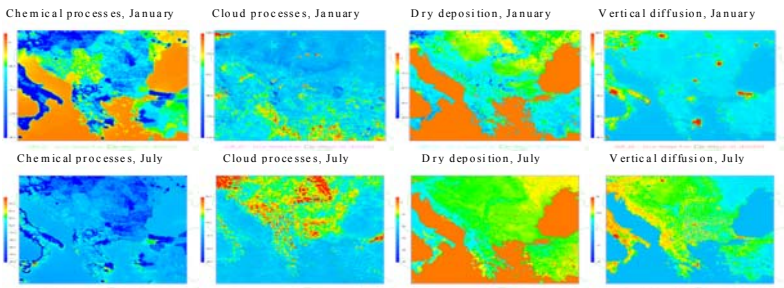


Figure 2. Plots of the contribution [µg/m<sup>3</sup>] of Chemical, Cloud, dry deposition and Vertical diffusion processes to the formation of O<sub>3</sub> at 16h of a "typical" day in January and July.

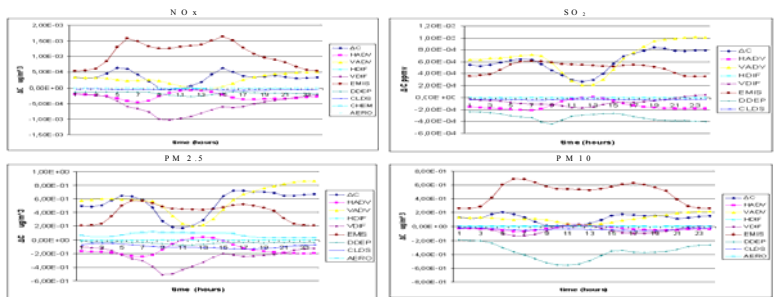


Figure 3. Plots of the diurnal course of the contributions [µg/m<sup>3</sup> and ppm V] of the different processes to the formation of NO<sub>x</sub>, SO<sub>2</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> and the resulting hourly concentration change  $\Delta C$  for a "typical" day in January.

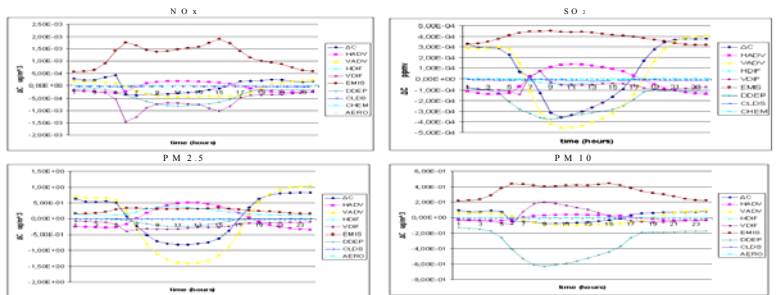


Figure 4. Plots of the diurnal course of the contributions [µg/m<sup>3</sup>] of the different processes to the formation of NO<sub>x</sub>, SO<sub>2</sub>, PM<sub>2.5</sub> and PM<sub>10</sub> and the resulting hourly concentration change  $\Delta C$  for a "typical" day in July.