REGIONAL MODELLING OF EPISODES OF EXCESSIVE AMBIENT AIR POLLUTION ON THE AREA OF SLOVENIA

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1. INTRODUCTION

The most important source of information on ambient air quality resp. on the concentrations of pollutants in ambient air are measurements, yet with them we can monitor air quality only on a limited number of measuring points, often without the possibility to define the causes of occurrence of excessive ambient air pollution. These disadvantages of measuring networks can be overcome with the assistance of models which can be used much wider and are dependent also of the art of the model (Žabkar et al. 2012a). In this paper we will limit ourselves to the modelling of ambient air quality with complex numeric models, that encompass a mathematical description of all important meteorological, physical and chemical processes which connect emissions and final concentrations of various ambient air pollutants. By such numeric tools for modelling ambient air quality it is possible to define the reasons and to evaluate the consequences of pollution, study various scenarios, predict air quality and anticipate the efficiency of various measures for improving ambient air.

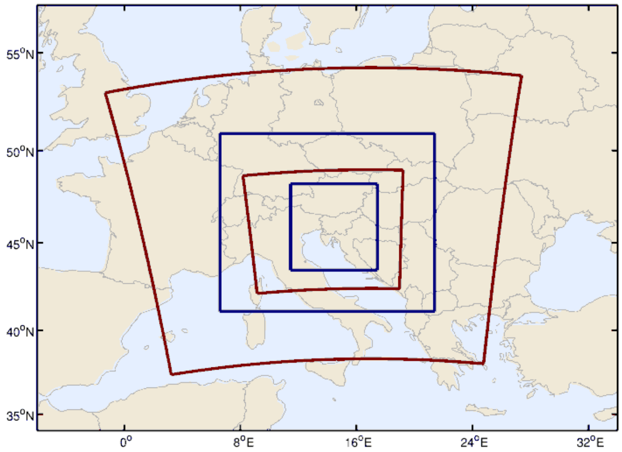
In the paper we analyse the main characteristics of a time and spatial development of certain episodes when in Slovenia exceeded concentrations of pollutants in ambient air were measured. This is done with the help of modelling results with the complex numeric models WRF/Chem (Skamarok et al. 2008) and ALADIN/CAMx (Team, 2003; ENVIRON, 2011). We are discussing summer episodes of high ozone concentrations and winter episodes of exceeded concentration of particles in ambient air. We wish to exemplify the mechanisms which lead to the occurrence of pollution episodes and at the same time we are preparing a basis for a successful prediction of ambient air quality for Slovenia with the assistance of the stated numeric models. When evaluating the results of the models, equally important as the quantifiable match of the model calculations with the measures is a quality match which shows how the model calculations follow the actual dynamics of the studied episodes. In any case we need to be aware beforehand that in models it is not possible to describe all processes in nature with an arbitrary accuracy. Due to sometimes incomplete knowledge of complex processes and mechanisms, incomplete information about the initial state of the atmosphere and on the outline of the model area and because of limited computer capabilities it is necessary when using models according to their purpose to seek a balance between available information on the state of the atmosphere and surface, complexity of the processes' description in the model and the speed of the numeric calculations.

2. CHARACTERISTICS OF THE APPLIED MODELS

2.1 WRF/Chem

The WRF/Chem (Grell idr., 2005) is a merged version of the meso-meteorological model WRF (Weather Research and Forecast; Skamarock et. al. 2008) and thus it enables a simultaneous modelling of meteorological processes, chemical transformation and the transport of pollutants in each calculation step. At the moment, coupled modelling represents the most advanced approach towards description of complex interoperation of physical and chemical processes in the atmosphere. Besides influences of physical atmospheric parameters on the dispersion and chemical transformations of pollutants such a model also incorporates influences of pollutants in the atmosphere on different meteorological resp. physical processes in the atmosphere such as radiation balance, cloud formation and precipitations. A combined calculation method on one side enables that when calculating transport and chemical conversions of pollutants we can include a maximum number of information on the meteorological state of the atmosphere and simultaneously in the calculations of the meteorological forecast we can also include the influence of pollutants in the atmosphere.

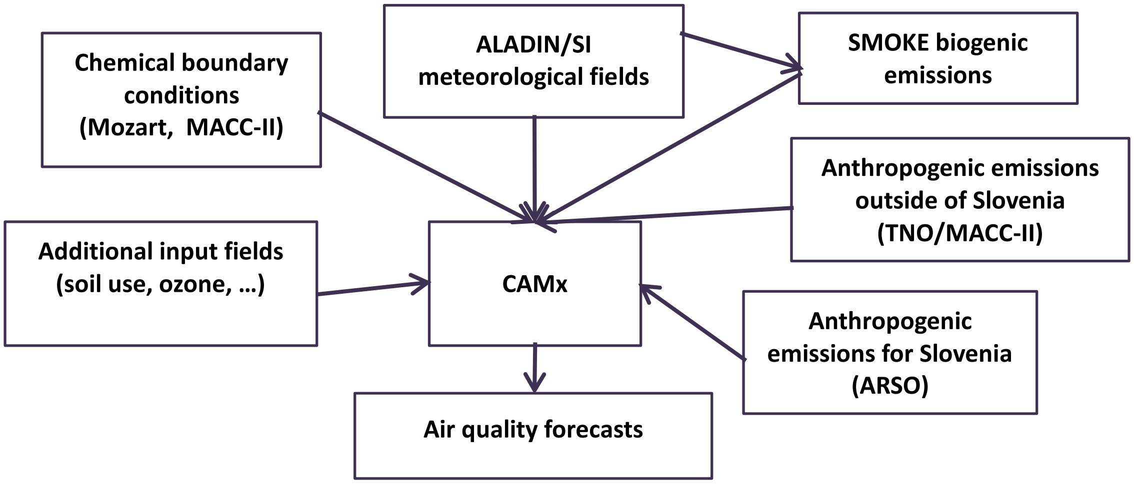
Thus, with the model WRF/Chem we are simulating meteorological activities and at the same time chemical conversions and the transport of pollutants within a specified area. In our simulations the starting and border meteorological conditions derive either from fields of the European Centre for Medium-Range Weather Forecasts, ECMWF, or from fields of the Global Forecast System (GFS) of the NCEP-model. We always use a nesting procedure, where we run the model through more nested calculation areas (as is the case in picture 1). Our area of interest is always the inner calculation area with the highest resolution, yet we need calculations in the outer border calculation areas in order to ensure good chemical and meteorological border conditions on the borders of the inner area. Chemical border conditions (pollutants' concentrations) on the borders of the outer calculation area are gained from calculations of the global MOZART-model (Model for Ozone and Related chemical Tracers), for the starting conditions we use results of a preliminary simulation. Anthropogenic emissions (7 primary pollutants, 5 sub-groups of particles and 48 sub-groups of volatile organic compounds) for the area of Slovenia are calculated from similar annual data on emission sources for the year 2009 (Source: Slovenian Environmental Agency), while for the areas outside of Slovenia we use the MEGAN-model (Model of Emissions of Gases and Aerosols from Nature) within WRF/Chem. This included in its calculations also the use of soil and current local meteorological conditions. For evaluations of sea aerosols and dust rise we use modules combined with the WRF-Chem model, which also enable the evaluation of these emissions in correlation to land use and current local meteorological conditions. In order to calculate the chemical conversions among gaseous pollutants we use RADM2 and for particles the MADE/SORGAM chemical mechanism. We chose the parametric schemes in the model for micro-physical processes, for an evaluation of flows between the surface and atmosphere, for turbulences in the planetary border layer, for photolysis and the parametric convective cloudiness in a way where the meteorological model also considers the presence of pollutants in ambient air. The vertical axis divides the atmosphere between 42 and 51 levels, whereas the levels are substantially more dense near the surface of the area of the planetary boundary layer (e.g. around 20 levels in the lower 2 km).



Picture 1: Modelling areas: blue areas indicate examples of nesting as used in the experimental operative forecast with the WRF/Chem model (available on: http://meteo.fmf.uni-lj.si/onesnazenje). The outer zone has a resolution of 11.1 km and 150x100 network points, the inner zone has a resolution of 3.7 km and 181x145 network points. Red indicates examples of modelling when the ALADIN/CAMx model of the Slovenian Environmental Agency (ARSO) is used for modelling. The outer zone has a resolution of 13.2 km and 145x135 network points, and the inner zone a resolution of 4.4 km and 185x167 network points.

2.2 ALADIN/CAMx

Some examples of results which are presented below were calculated with the ALADIN/CAMx model system (Žabkar et. al. 2012b). This is the model system of the Slovenian Environmental Agency (ARSO), where the (photo)chemical-transport model CAMx (Comprehensive Air quality Model with extensions; ENVIRON, 2011) uses meteorological fields of the ALADIN model (Team, 2003) in a time interval of 1 hour. The US EPA – Environmental Protection Agency authorised the CAMx model for ozone and particle modelling in various time and space scales. CAMx needs for its calculations meteorological fields, which in our case are forecasts of the ALADIN model and some other input fields, mainly time-variable emission fields, different geographical variables and data on the total volume of ozone in the air. In order to calculate chemical and photochemical conversions in the model we use the SAPRC99 (Carter, 2000) chemical mechanism, which encompasses 114 different chemical compounds respectively compound groups, among them 16 radicals, 22 particle groups and 217 chemical reactions. The implemented model configuration includes 34 vertical levels (up to a height of 14 km). The model CAMx receives the starting chemical conditions from the previous operation while for the border chemical conditions results of the MOZART model (MACC-II project) are used. As with the configuration of the WRF/Chem model also the ALADIN/CAMx model in Slovenia uses similar data on annual emission sources for the year 2009 (Source: ARSO), while for areas outside Slovenia TNO/MACC emissions are used. The SMOKE model (Sparse Matrix Operator Kernel Emissions) is used for the calculation of biogenic emissions. A simplified schematic of the ALADIN/CAMx model system is shown in Picture 2.



Picture 2: Simplified schematics of the ALADIN/CAMx model system.

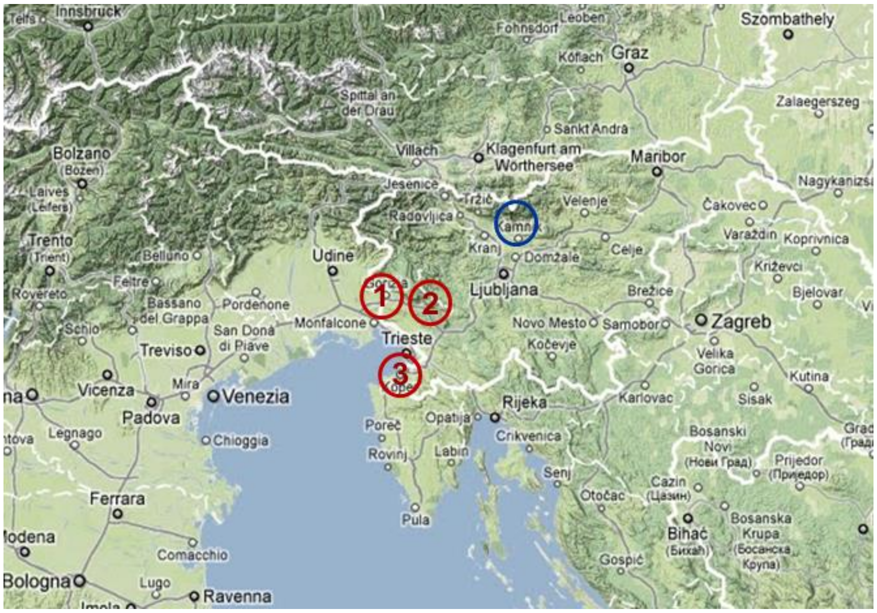
3. EPISODES OF EXCEEDED OZONE CONCENTRATIONS

Tropospheric ozone is a secondary pollutant and it is formed in complex (photo)chemical reactions from primary pollutants, mostly from nitrogen oxides (NO_x) and volatile organic compounds (VOC). High concentrations of ozone in ambient air occur, when in the air masses sufficient total pollution is accumulated (also the ratio between the volume of NO_x and VOC is important) and if at the same time weather conditions are favourable for the formation of ozone. For an accumulation of a sufficient amount of pollutants in ambient air normally a few days of favourable weather conditions are enough. For an efficient formation of ozone also high ambient air temperatures and adequate sun radiation are necessary, which is the reason why the highest concentrations occur in the warmest months. If, however, the winds are not strong enough and a thinning of the pollution is less effective, ozone concentrations can reach very high values in such circumstances. When specifying the locations of the highest concentrations of ozone we need to consider also that time is needed for ozone formation out of emissions of primary pollutants, thus typically the highest concentrations of ozone are occurring in wind directions somewhat outside of city centres respectively away from bigger emission sources.

So far studies of ozone episodes were restricted only to episodes when on one or more measuring points in Slovenia the daily maximum ozone concentrations were exceeded, i.e. when the highest measured daily concentration in Slovenia was higher than 180 $\mu g/m^3$. Further on we summarise some main characteristics of these episodes with extremely high daily maximum ozone concentrations. More problematic than the exceeded daily maximum values are exceeded target values for a time span of 8 hours (120 µg/m^3) which were measured much more often. A detailed analysis of episodes with exceeded 8-hour target values might reveal additional features of their dynamics, as we were not able to ascertain them with exceeded daily maximum values.

Table 1: Number of days with measured daily maximum ozone values above 160 µg/m^3 for the last three years.

Measurng point \ year	2010	2011	2012
Nova Gorica	13	16	22
Koper	7	4	12
Otlica	13	15	12
Krvavec	8	1	11
Ljubljana	-	2	4
Maribor	-	-	-
Celje	1	2	4
Murska Sobota	2	2	1
Trbovlje	2	2	2
Zagorje	1	-	1
Hrastnik	4	2	3
Iskrba	-	1	2

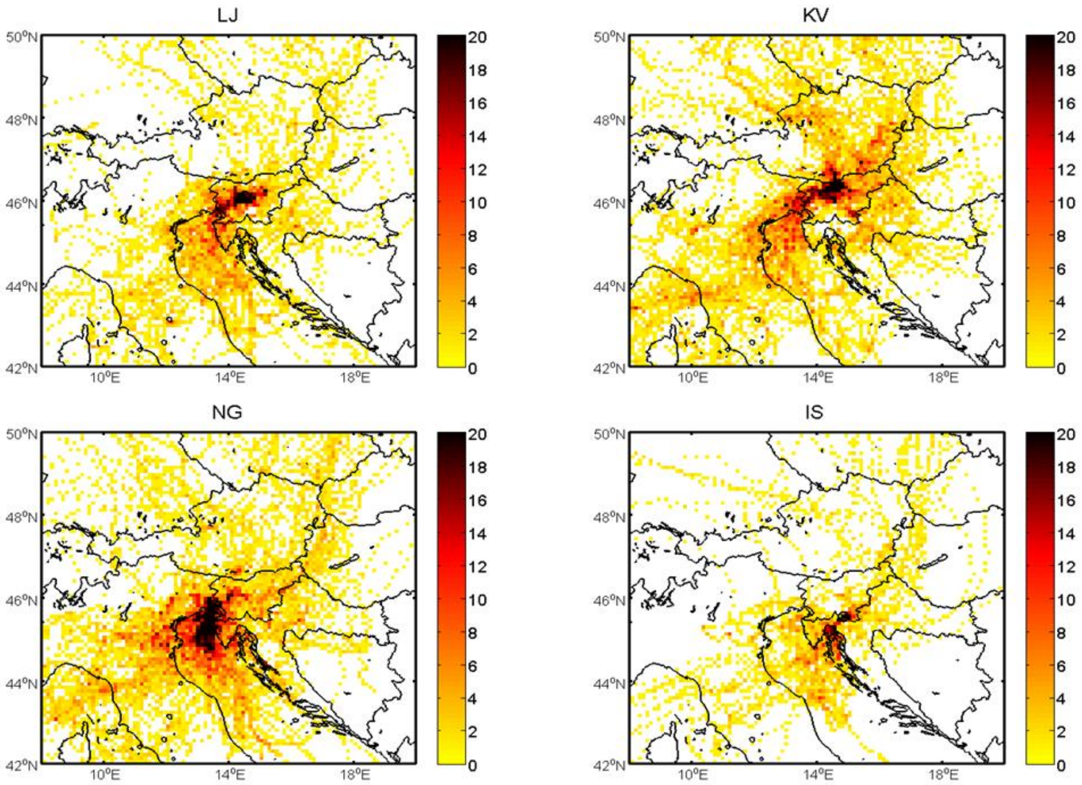


Picture 3: Locations of measuring points where the measured daily maximums of ozone were most often exceeded. In red are the following measuring points: Nova Gorica (1), Otlica (2) and Koper (3), in blue is the measuring point on the mountaintop Krvavec.

In Slovenia most often daily maximums of legal thresholds are exceeded in the coastal region (measuring points: Koper, Otlica, Nova Gorica) and on the highest situated measuring point in Slovenia on the mountain of Krvavec. Exceptionally very high concentrations of ozone may occur also in urban locations in the inner parts of the state and even at the measuring point Iskrba, which is remote from big emission sources and intended for measuring background ambient air pollution in Slovenia (table 1, picture 3).

3.1 Analysis of trajectories

Slovenia is a small country and because of this the question arises to what extent high ozone concentrations in our state are a consequence of inherent emissions respectively what role does a cross-border transport of polluted air masses play within high levels of pollution. This question is even more relevant with exceeded ozone levels in the coastal region, as some research has shown that the Po plain is the source of polluted air masses in the Alpine area (e.g. Kaiser et al., 2007). In order to check whether polluted air masses from the Po plain represent an important influence on the daily exceeded ozone levels in the coastal region we conducted an analysis of the direction of air flows (Trajectory, Žabkar et. al., 2008). With it we assessed the origin of the air masses for those days when high ozone concentrations were measured. Although only an analysis of the trajectories is not enough for a final conclusion, as beside the transport it does not enable a discussion about the influence of other processes (chemical conversions, dry depositions, flushed/rinsed precipitations, turbulent diffusions), still the trajectories can show us a possible connection between high ozone concentrations in Slovenia and areas outside of Slovenia and which air masses flow through before reaching our areas.



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Picture 4: Numeric density of “polluted” trajectories backwards for episodes when the 3-hour ozone concentration at the final point of the trajectory (measuring point) was higher than 160 μg/m^3. The results show the measuring points in Ljubljana, Nova Gorica, Krvavec and Otlica.

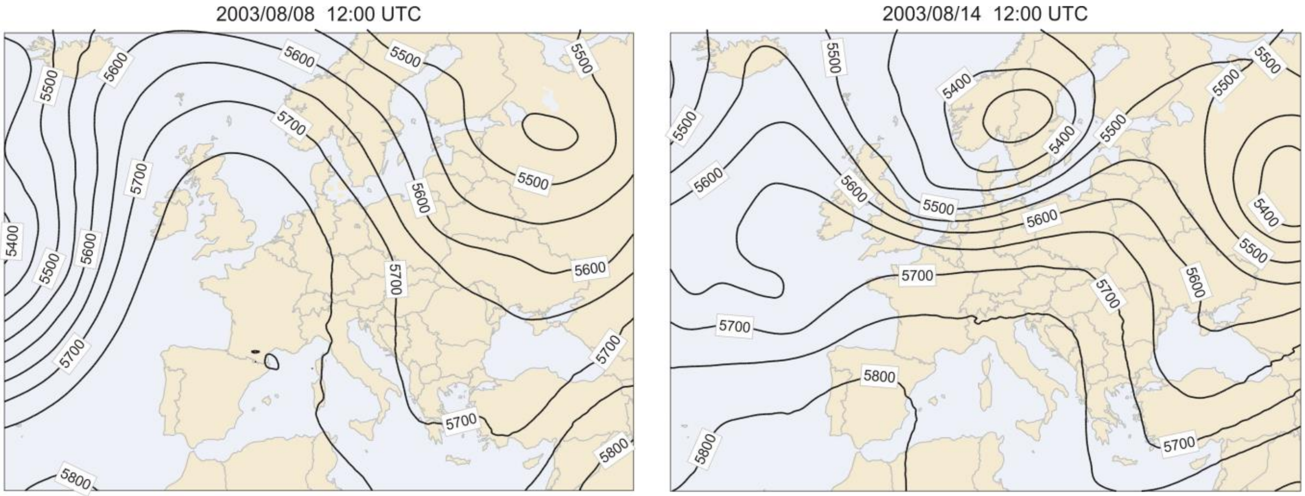
For the calculation of three-dimensional (3D) trajectories we used meteorological fields calculated with the ALADIN model (Team, 2003) with a horizontal resolution 9.5 km and 37 vertical levels. The trajectories were calculated with the FLEXTRA model (Stohl et. al., 1995, Stohl and Seibert, 1998) for the warm annual half (April-September) of the years 2003 and 2004. The final points of the trajectories were specified 50 m above the model topography, their “arrival” times to the measuring point were set to every 3 hours (00, 03, …21 UTC) The length of the trajectories was 96 hours, or applicable shorter in cases when the trajectory “left” the calculation area earlier.

The example of results of the trajectories’ analysis is presented on picture No. 4 and it shows the numeric density of the trajectories back for some measuring points for times when the measured 3-hour concentration of ozone in the time of the trajectory arrival to the measuring point was above 160 μg/m^3. The analysis of these “polluted” trajectories showed as the most probable source of ozone polluted air masses for Slovenia the area of the northern Adriatic Sea. As besides ships there is no recognisable source of primary pollutants, potential sources of pollution are considered to be sources from the coastline – on the shores of the bays of Triest and Rijeka are significant industrial sources (factories, two harbours, thermal power plants, a refinery), similar to the western coastline of the Adriatic Sea on the Italian coast. A direct influence of the emissions from the Po plain on the ozone in Slovenia could not be confirmed with the analysis of trajectories. We need to be aware that fields of numeric density of the polluted trajectories only offer information on the direction of potential pollution sources and not necessarily on their location. For example we need to consider that a high numeric density of polluted trajectories above a certain area is also a consequence of air canalisations (windward/downwind area of the measuring point). For that purpose fields of average 3-hour maximum concentrations of ozone on all measuring points connected with air masses were significantly higher. Here we are talking about air masses coming from above the northern Adriatic Sea and the coastline. Only in the town of Nova Gorica a maximum could be recognised on the area of the Po river mouth at the Adriatic Sea, which indicates a probability of emission influences of the Po plain on that measuring point. On other measuring points no signs of an increased influence of emissions from the Po plain could be noticed. Nonetheless, emissions from the area of the Po plain still significantly influence ozone concentrations, e.g. by contributing to a broader “island of polluted air”, which form above the broader area of the northern Adriatic Sea.

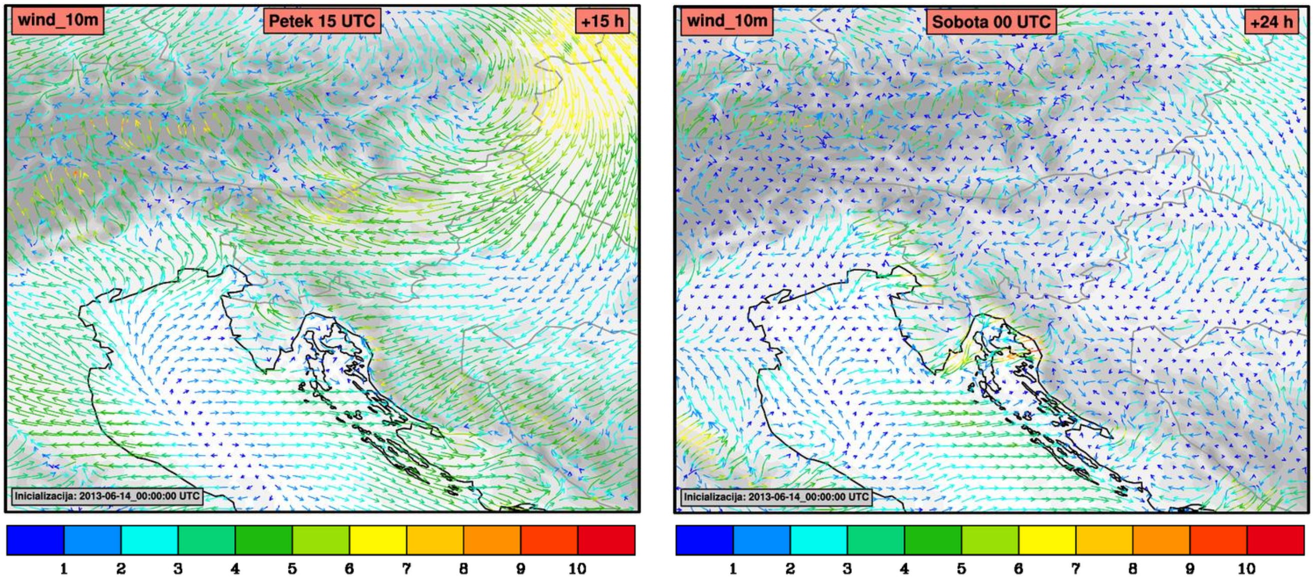
3.2 Modelling results

We researched with the help of modelling the occurrence of the Adriatic Sea as a “source” of ozone-polluted air masses for Slovenia. It could be possible that the polluted masses are actually accumulating above the sea, or the Adriatic Sea is functioning only as an apparent pollution source. In this case important intakes of pollution from coastal areas into the air masses may occur on their path.

We modelled the development of a certain number of ozone-episodes, when the measured daily maximum values exceeded the allowed values. Most of the simulations were made with the WRF/Chem model, and some newer simulations with the ALADIN/CAMx model. Despite the fact that the simulations were made for different episodes (similarity/difference criteria were duration, expansion, paths of 3D-trajectories) and with different models, the results showed that the treated episodes have some common characteristics. Thus, the development of the episodes through time, when at the coastal measuring points the highest ozone-concentrations occur, consist of three typical phases and are closely tied to the development of a meteorological situation in synoptic scales.

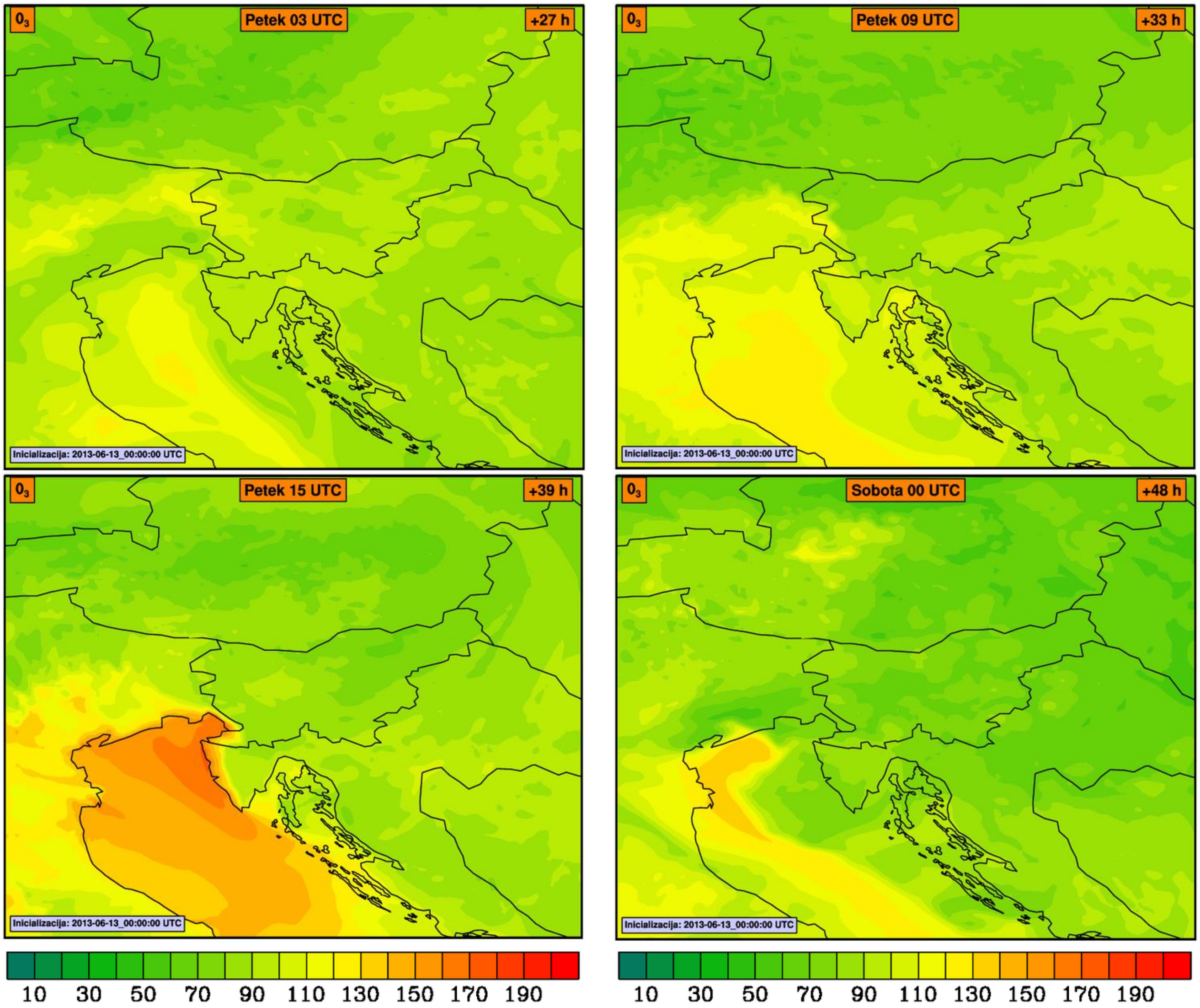


Picture 5: Geopotential field on a 500 hPa area for August, the 8th and 14th 2013 at 12 UTC.



Picture 6: Wind 10 m above surface in m/s on June, 13th 2013 at 15 UTC (left) and June, 14th 2013 at 00 UTC (right). Results of the WRF forecast are shown (available at: http://meteo.fmf.uni-lj.si/vreme).

In the first part of the episodes the synoptical situation is defined by the gradual phase of the geopotential on an area of 500 hPa, which is situated westward of Slovenia (e.g. in Pic. 5, left). In this time Slovenia is located on an area of high air-pressure at the surface, while in the higher air layers NW winds blow, which are blocked in the lower atmospheric layers by the Alpine barrier. Partial and foremost above NE Slovenia an air mass engulfment is occurring from NE, but mainly above lower air layers above Slovenia where there are no predominant general flows. The main driving forces for air in this period are occurrences in the mesoscale (local hillside winds, valley winds, coastal winds, heat islands above cities etc.) At coastal areas in this time typical thermic induced winds occur, which in the case of the northern Adriatic Sea are gathering to continue as hillside winds in the Alpine foothills and in NW remains of the Dinaric mountains. At daytime we have a predominant air direction through the region of Friuli and further towards the mountain peaks and at night-time in the opposite direction from the hills towards the sea (picture 6). The so created circulations or even re-circulations of air masses above the test area can be persistent for some time which enables them to accumulate inside high concentrations of pollution from emission sources. Simultaneously during this episode phase the weather conditions on the Mediterranean are also quite favourable for the formation of ozone and therefore the final concentrations of ozone can reach very high values. Slovenian areas eastern from the Alpine-Dinaric barrier are in this part of the episodes more or less separated from the Mediterranean air masses. For the rest of Slovenia the first part shows lower episodes of concentration peaks, and also exceeded 8-hour values of ozone can occur. Picture 7 shows a case of time development of ozone for a day in the accumulation phase of pollution above the Adriatic Sea.



Picture 7: Ozone development in the air layer at surface on June, the 13th 2013. Shown are results of the experimental ozone forecast with the WRF/Chem model (available at http://meteo.fmf.uni-lj.si/onesnazenje).

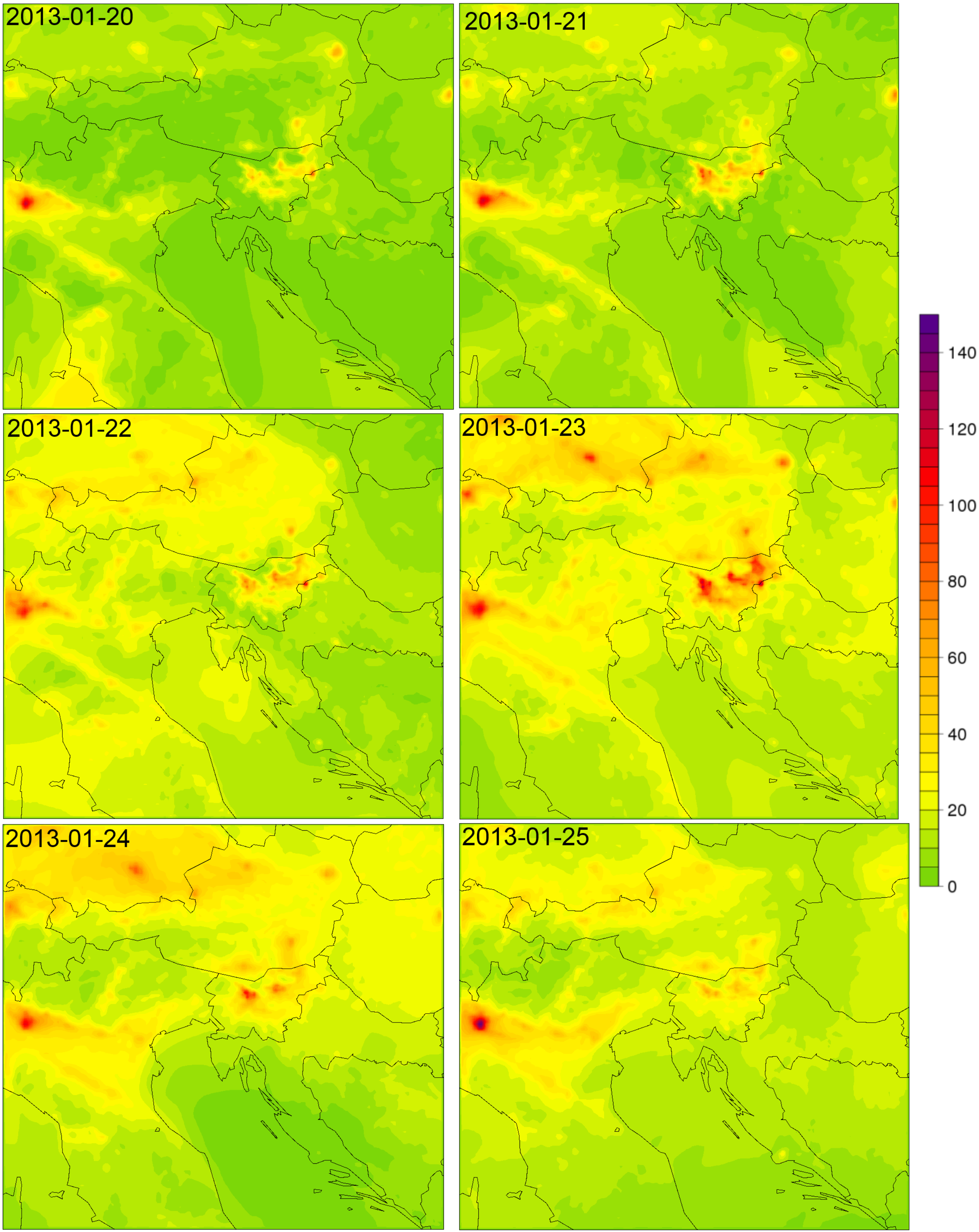
In the second part of the episodes connected with transport of the geopotential ridge on 500 hPa areas in eastern direction, south-west winds occur in Slovenia, where the polluted air masses above the Adriatic Sea are moved towards and above Slovenia. As the pollution extent above the Adriatic Sea can be substantial, it can happen, that in these days also at some places of central Slovenia higher ozone values are measured. This also explains the highest occurrence of high ozone concentrations for trajectories from a SW direction also for the measuring points in central Slovenia.

The episodes are normally interrupted by fronts, after which ozone concentrations fall considerably. A drop in ozone concentration occurs in case of intense front precipitations (light precipitations or scattered showers of rain and storms do not clean the atmosphere entirely), which also clean the atmosphere, and in cases of cooling of the lower troposphere and when the intensity and direction of the air flow above Slovenia changes.

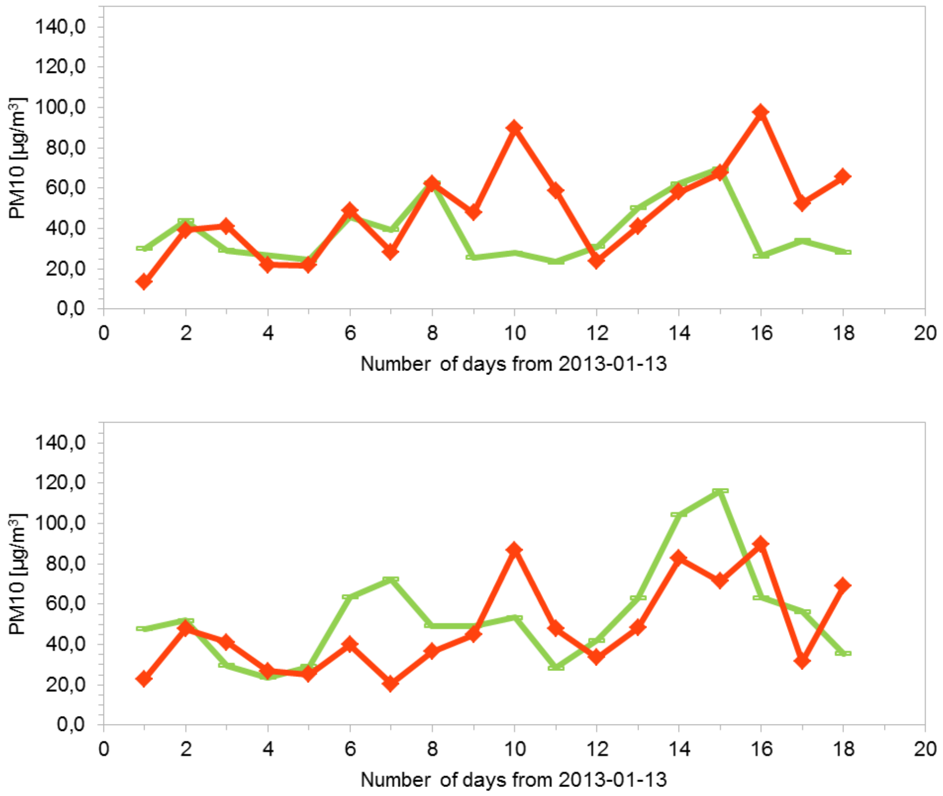
4. EPISODES OF EXCEEDED PM-CONCENTRATIONS

Although high concentrations of airborne particles in colder (winter) months is a general (European) problem, Slovenia's location underneath the Alps and the relief structure of the landscape additionally contribute to high concentrations of PM. As in moderate geographical longitudes west-winds dominate and Slovenia lies eastern from the Alpine arc, therefore on the lee side, so winds in our area are not strong in general. There is much calm or only weak winds, which is not favourable for thinning pollution. Because of the diverse relief structure general winds are directed according to relief, and there are also regional coastal and hillside winds.

Most of Slovenia's surface is hilly and most of villages, towns and arterial roads (most of the emission sources) are situated in valleys and basins. Valleys and basins are in their relief of concave forms and thus there and in the surrounding hillsides a very characteristic air flow is occurring (Rakovec et al. 2012). During the day the sunny hillside warms up the most and here air begins to rise and daily upward hillside-winds occur. With these winds the air under the hillsides changes and polluted air is raised from the plains upwards on the hillsides. In the afternoon the surface begins to cool down and downward airflows start where cold air from the hillside flows into the valleys and basins and they create areas of cold air. Night hillside winds are in general weaker yet mostly in the colder part of the year (short days, long nights) they can last most of the day. If the valleys are fairly closed (the basins are closed anyway), then this cold air accumulates and during long nights basins and valleys can fill up entirely with cold air. Thus, the floor of a basin or a valley is the coldest and higher layers are warmer, and this is called inversion. In such circumstances pollution thinning in the air via turbulence diffusion may completely stop. If during the day there is no thermic or dynamic degradation of the temperature inversion, such a condition can endure even more days or weeks (in the colder half of the year). All emissions into cold air in the basin remain closed in the basin, as the inversion prevents exchange of air from higher layers. At the same time this also means that the volume which for days is fed from chimneys, exhaust pipes, and industrial ventilation systems etc. is limited. A consequence is of course exceeded concentrations of pollutants, especially of particles, in basins and valleys.



Picture 7: Average daily concentrations of PM10 in μg/m^3 for 6 consecutive days within an episode in January 2013. The shown results were calculated with the ALADIN/CAMx model system.



Picture 8: Comparison of simulated (red, ALADIN/CAMx model) and measured (green) average daily concentrations of PM10 in µg/m^3 for Ljubljana (upper chart) and Maribor (lower chart) within the second half of January 2013.

In basin areas cold air may develop and create weak, isolated air movements. Sometimes these are so weak, that even the most sensitive wind meters can hardly recognise them. On the areas where downward hillside flows are joining a slight compensating air-rise towards the top of the area of cold air occurs and underneath the hilltop they flow out and on the hillside down again. These weak air movements with regional models (resolution some km) of course cannot be simulated but the possibility is fairly good to simulate the phenomenon of temperature inversions and with it we can connect increased concentrations of pollutants in basins and valleys. Picture 7 shows an example of simulated concentrations for PM10 particles for the area of Slovenia and its surroundings for 6 consecutive days in January 2013, when measured PM10 concentrations in Slovenia were very high. Picture 8 shows for this period (second half of January 2013) a comparison between model results and measurements for two measuring stations, Ljubljana and Maribor. We can see that the simulated concentrations of particles from January, the 20th till January, the 23rd rose gradually from day to day, whereas the highest concentrations in Slovenia occur in settled basins and valleys of the continental area of the state, while in the coastal region the simulated (and measured) concentrations of particles were lower. From January, the 24th onwards the simulated concentrations of airborne PM10 fell; similar results are evident also at the measurements. A detailed analysis between the simulated and measured concentrations according to individual measuring stations showed for this episode that the model followed well the measured concentrations; according to expectations the model could not simulate concentrations with a high quality respectively the differences in PM10 concentrations between individual valleys and basins. For such purpose the model calculations have a too low spatial resolution, next to it there are too many uncertainties at the input data of the model (within the emission data, starting and border pollutant concentrations, uncertainties in the model regarding the description of various processes etc.).

5. CONSLUSION

The formation efficiency of tropospheric ozone depends on weather conditions. The highest ozone concentrations in the surface air layers occur at calm summer anticyclone situations, when outside temperatures are high and there is much sun radiation. As the formation of ozone from primary pollutants takes time, the highest ozone concentrations are typically occurring in wind direction somewhat away from city centres respectively from bigger emission sources. In Slovenia daily ozone maximums are most frequently exceeding the legal thresholds in the coastal region and on the higher elevated measuring station on the Krvavec mountain. Simulations of episodes with high measured daily ozone maximums showed common characteristics between the episodes. Their time development is made of three distinct phases. In the first part of the episodes the main transport drive are occurrences in the mesoscale (local hillside, valley, coastal winds, heat islands above cities etc.). On the coastal region circulations and even re-circulations of air masses occur, which enable that here from emission sources of these areas great pollution masses accumulate. Simultaneously, during this episode phase weather conditions in the coastal region are usually good for ozone formation, thus final ozone concentrations can reach very high values. In the second part of the episodes above Slovenia south-western winds occur which transport the polluted air masses from above the Adriatic Sea towards and over Slovenia. The episodes are usually interrupted by passing weather fronts, after which ozone concentration falls rapidly.

As a difference to ozone particle concentrations are the highest in winter months, when the exceeding of legal thresholds occurs in urbanised basins and valleys inside Slovenia, while in the coastal region less exceeding occurs. Next to emissions also poor aeration of basins and valleys contribute to high concentrations of particles in the diverse inner parts of Slovenia and they are often connected to occurring temperature inversions. These inversions can in winter months endure also for many days or even weeks and in such cases all emissions in a basin stay enclosed. Although the used regional models cannot exactly simulate the quantitative progression of particle concentrations in individual basins, we can simulate quite well the general dynamics and time development of episodes of high concentrations of particles in the area of Slovenia.

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