A model study of ground level ozone pollution in the High Tatras Mountain region

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Abstract: Air pollution, especially ground level ozone (O_3), negatively influences sensitive ecosystems of the Tatra National Park (UNESCO Biosphere reserve). The aim of this work is to analyse the role of long-range transport in extremely high ozone air pollution situation over the High Tatras Mountain region. Summer O_3 episode observed during 12-14 August 2003 was modelled by coupled meteorological and photochemical mesoscale model MetPhoMod *(Perego, 1999)*. Standard meteorological data and O_3 concentration measured at fixed ground stations: Poprad - Gánovce, Stará Lesná, T. Lomnica - Štart, Skalnaté Pleso and Lomnický štít in vertical profile from 706 to 2634 m a.s.l. were used. Emissions and land cover type of model domain (20 × 18 km) were obtained from EMEP database and regional land use maps, respectively. Observed maximal O_3 concentrations were about 60-90% higher than the simulated values (considered as local O_3 forming potential). During the peak phase of investigated O_3 episode, the contribution of local independent emission sources to observed O_3 concentration was on average 37%. Achieved results indicate that long-range transport (from Western Europe) and descent of O_3 enriched air from high troposphere to ground level supported by high pressure system played more significant role in O_3 concentration increase than its local formation during the extremely high O_3 events in the High Tatras Mountain region.

Key words: ground level ozone, episodes, temporal and spatial variation, complex topography, High Tatras Mountain, emissions, meteorological factors, photochemistry, model MetPhoMod

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1. Introduction

Air pollution is a serious problem in the High Tatras Mountain (*Fleischer et al., 2005*). Both north high altitude areas of the Tatra National Park (UNESCO Biosphere reserve) and southwest low landed vicinity of the Slovak capital city Bratislava are localities most affected by the ground-level ozone (O_3). At these places, air quality standards were frequently exceeded during O_3 episodes in August 2003 (*Bičárová et al., 2005*). In this paper, the summer O_3 episode observed during 12–14 August 2003 in the High Tatras Mountain region is investigated using model MetPhoMod (*Perego, 1999*). Model study of O_3 pollution enables to analyse local O_3 forming potential and the contribution of emission sources to maximal O_3 concentrations. Recent O_3 concentration in Slovakia is substantially more affected by long-range transport and by climatic changes than by national strategy application for reduction of O_3 precursor emissions (*Hrouzková et al., 2004*). O_3 simulation during the August 2003 heat wave performed by chemistry-transport model CHIMERE also shows the significant role of transboundary transfer of O_3 and its precursors in Europe (*Vautard et al., 2005*). MetPhoMod model simulation of O_3 concentration from local emission sources at model domain and comparative analysis using measured values of O_3 concentration is one way of specifying the role of long-range transport to air pollution in the High Tatras Mountain region.

2. Materials and methods

2.1. MetPhoMod model description

MetPhoMod (Meteorology and Photochemistry Model) is the eulerian, three-dimensional, mesoscale model for simulation of summer smog over very complex terrain under fair weather conditions *(Perego, 1999)*. It is a single program that includes modules for air motion, turbulence, radiation, ground-atmosphere interactions, gas phase chemistry, and deposition. The model uses a cartesian grid. Complex topography is considered by dividing grid points into two categories: normal and underground. Transport is calculated using the PPM (Piecewise Parabolic Methods) based transport scheme. The nonhydrostatic pressure is evaluated by solving an elliptic equation derived from the mass continuity equation. The turbulence module implements the k-ε turbulence closure scheme with an implicit solver. The program includes a chemical interpreter and predefined input file of chemical equations for the RACM - Regional Atmospheric Chemistry Mechanism *(Stockwell et al., 1997)*. MetPhoMod does not include modules for clouds, aerosols and heterogenous chemistry. A grid of rectangular cubes represents the modelling domain. All values are stored in the centre of the cube. The dynamics then are solved with the method of *Rhie and Chow (1983)*, in the form proposed by *Clappier (1998)*. The solution procedure strictly keeps mass consistency. The chemical equation system is solved with a technique based on separation into fast and slow species *(Gong and Cho, 1993)*. The fast species are solved with an implicit, the slow ones with an explicit integration step. The MetPhoMod software consists of a single UNIX executable file (`http://www.giub.unibe.ch/klimet/metphomod/`) and is handled with common UNIX-commands. The netCDF (network common data format) binary data format defined by UNIDATA/UCAR (`http://www.unidata.ucar.edu/`) is used for input and output files formulation.

2.2. Model domain

The High Tatras Mountain region area of size 20 km × 18 km was used as the model domain. Selected area extends from Svit in the West to Kežmarok in the East and from Lomnický štít in the Northwest to Poprad basin in the Southeast (Fig. 1). Model domain includes fixed ground stations in vertical profile: Poprad - Gánovce (H = 706 m a.s.l., $\varphi = 49°02'$N, $\lambda = 20°19'$E); Stará Lesná (H = 810 m a.s.l., $\varphi = 49°09'$N, $\lambda = 20°17'$E); T. Lomnica - Štart (H = 1200 a.s.l., $\varphi = 49°10'$N, $\lambda = 20°15'$E); Skalnaté Pleso (H = 1778 m a.s.l., $\varphi = 49°11'$N, $\lambda = 20°14'$E); Lomnický štít (H = 2634 m a.s.l., $\varphi = 49°12'$N, $\lambda = 20°13'$E). Slovak Hydrometeorological Institute (SHMI) provides meteorological and air pollution measurement at stations in cooperation with the Geophysical Institute of SAS (GPI) and the Research Centre of the Tatra National Park (RC TANAP). Standard meteorological parameters *(Ostrožlík, 2004)* and O_3 concentration measured by adjusted UV absorption ozone analysators are available from all stations *(Bičárová et al., 2005)*. More anthropogenic emissions produce stationary (semi-urban, industrial) and mobile sources around the towns Poprad and Kežmarok in comparison with sparsely populated high altitude mountain localities. On the other hand, forest vegetation at foothills releases important quantity of biogenic volatile organic compounds (BVOC) emissions. The model used horizontal grid of 21 × 19 cells at resolution of 1 × 1 km^2 with 22 vertical levels in elevated interval of altitude from 600 to 2700 m a.s.l. Digital topographic data and representative altitude for every cell were obtained by digitisation of High Tatras paper map (ratio 1:50 000) using software Didger and Surfer (`http://www.goldensoftware.com`). Digital Elevation Model (DEM) of investigated domain 20 km × 18 km × 3 km is shown in Fig. 2.

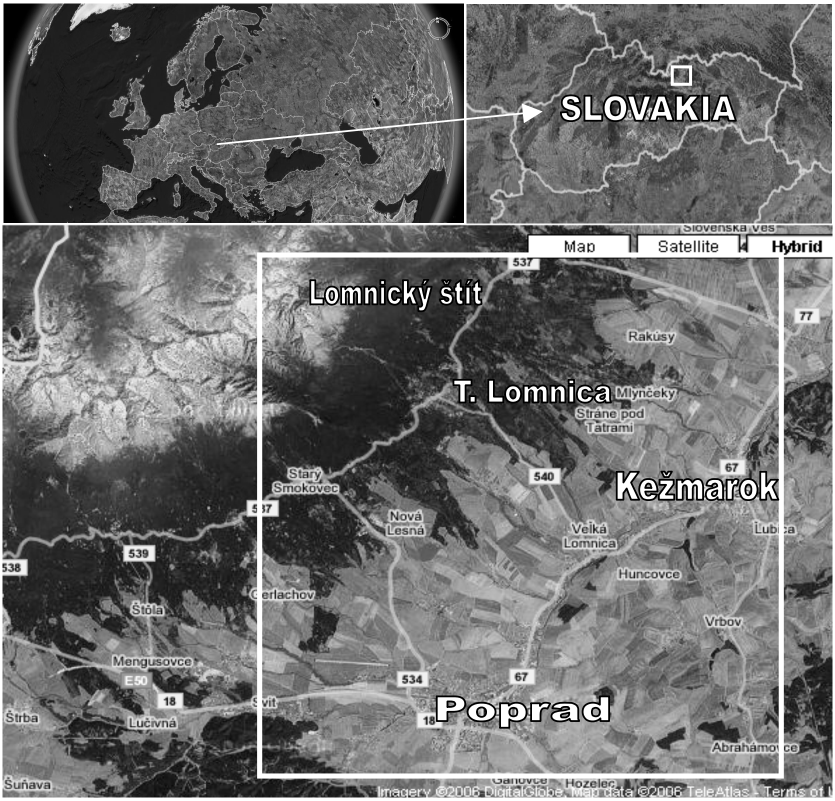

Fig. 1. Satellite images and borders of the High Tatras Mountain model domain (`http://geology.com/europe-satellite-images.shtml`).

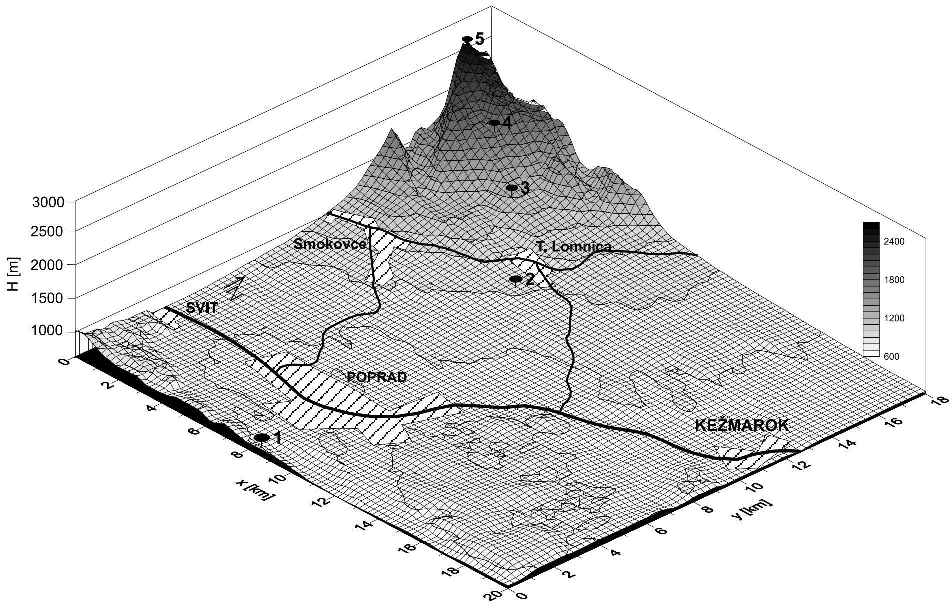

Fig. 2. Digital Elevation Model – DEM of the High Tatras Mountain model domain and location of O_3 and meteorological ground stations: 1 – Poprad - Gánovce, 2 – Stará Lesná, 3 – Štart, 4 – Skalnaté Pleso, 5 – Lomnický štít.

2.3. Input data – specification and validation

a) Static parameters
   Static parameters include relatively stable ground characteristic: ground roughness, the surface albedo, the ground heat capacity, the ground diffusivity, and the relative air humidity in ground pores, the evaporation resistance of soil and the shielding factor of plants. Parameters were defined in accordance with classification System of U.S.G.S (U.S. Geological Survey Land Use/Land Cover System). Land of the High Tatras Mountain model domain covers mainly vegetation of grass, shrubs, forest and mixed barren land of alpine rockies. There are also several semi-urban and rural settlements.

b) Dynamical parameters
   Dynamical parameters involve the meteorological data:

the wind direction, the wind speed, the air temperature, the air moisture, the pressure on top of domain and O_3 concentration. These data come from measurements at stations of the model domain. Hourly values are used to strengthen the model behaviour around the edges of vertical layers and on the top of domain. Fig. 3 illustrates the comparison between modelled and measured hourly values for air temperature, relative air humidity, wind speed, and wind direction at individual stations. Correlation coefficients show the good agreement between

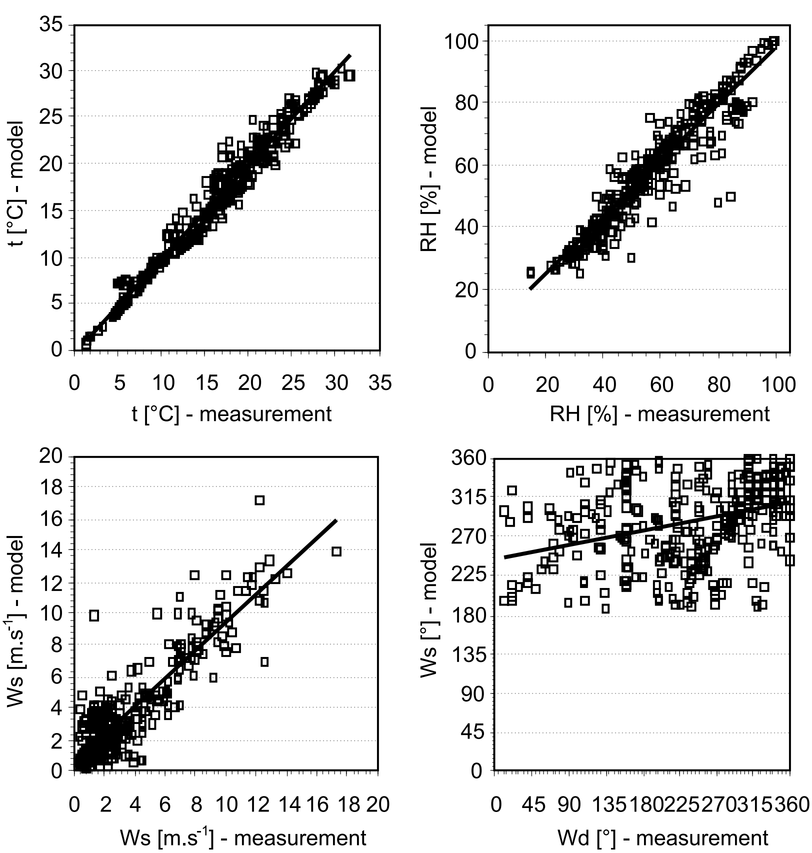

Fig. 3. Correlation coefficients (r) and relationship between modelled and measured values for air temperature t[°C]: r = 0.985, y = 1.012x – 0.478; relative air humidity RH[%]: r = 0.951, y = 0.915x +5.715; wind speed Ws [m s^{-1}]: r = 0.896, y = 0.892x + 0.498 and wind direction Wd[°]: r = 0.366, y = 0.187x +241.750 during the period 12-14 August 2003.

the experimental and modelled data except for very variable wind direction in the complex mountain area. Model calculates global solar radiation according to *Paltridge and Platt (1976).* The tight relationship between modelled and measured data at Stará Lesná (r = 0.936) indicates sufficient accuracy of the solar radiation model (Fig. 4).

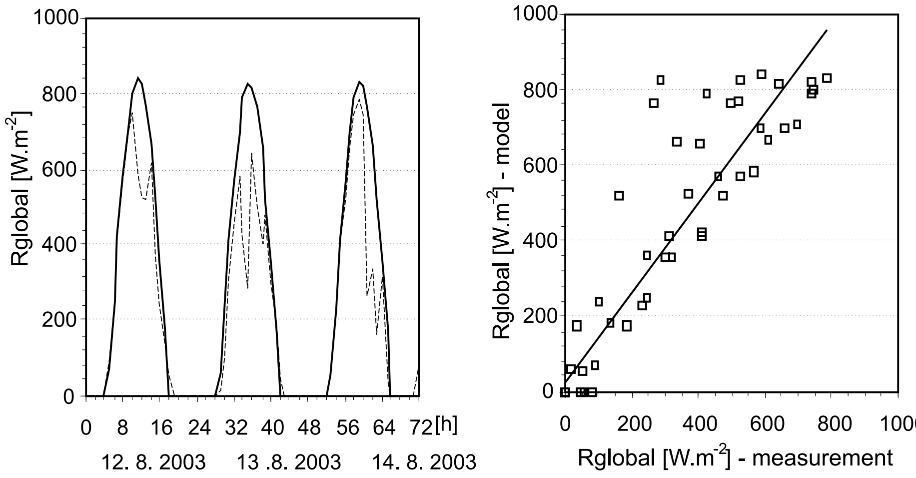

Fig. 4. Comparison between modelled (solid line) and measured values (dashed line) for global solar radiation Rglobal [W m^{-2}]: r = 0.936, y = 1.102x + 25.048 at Stará Lesná during the period 12-14 August 2003.

c) Emissions

Emission data of EMEP expert emission database WebDab *(Vestreng et al., 2005)* were applied to the model domain of the High Tatras Mountain region. WebDab database (`http://webdab.emep.int/`) enables access to national total, sector, and grided emissions for the listed areas, years, pollutants or activity classes and total or sector categories. The present EMEP grid domain consists of cells with resolution 50 × 50 km^2. Coordinates of EMEP grid square: X (i) = 78, Y (j) = 56 correspond to geographical coordinates of model domain: longitude 20.26°E, latitude 49.16°N. Temporal and spatial disagregation of annual (2003) EMEP grid square (78,56) emissions to grided emission values of model domain were performed for gaseous species CO, NO_x (NO and NO_2), SO_x and non-metane hydrocarbons NMHC. Composition and fractions of NMHC groups were assigned according to *Stockwell et al.(1990)*. BVOC emissions of model domain *(Bičárová and Fleischer, 2006)* were estimated by GLOBEIS-BEIS2 model *(Guenther et al., 1993)*. Fig. 5 shows the spatial distribution of average daily concentration of anthropogenic NO_x and biogenic ISO (isoprene) O_3 precursors at model domain obtained by the MetPhoMod. Maximal NO_x concentration corresponds with distribution of stationary and mobile emission sources as well as ISO concentration with forested area of model domain.

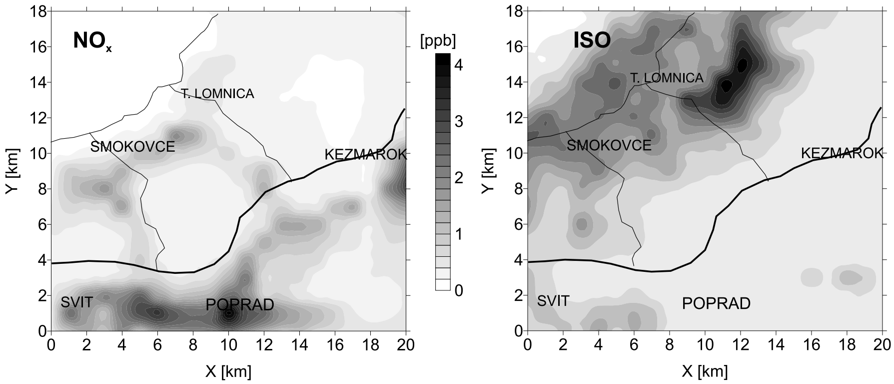

Fig. 5. Spatial distribution of modelled average daily concentration of anthropogenic NO_X [ppb] and biogenic ISO (isoprene) [ppb] ozone precursors produced by local emissions of the High Tatras Mountain domain for the period 12-14 August 2003.

3. Results and discussion

Extreme high O_3 concentrations were measured in Bratislava ($301 \mu g\,m^{-3}$) and at Lomnický štít ($195 \mu g\,m^{-3}$) in August 2003 in Slovakia *(Bičárová et al., 2005)*. Extraordinary O_3 pollution related to air masses forming above high-emission areas of West Europe during 1-12 August 2003 heat wave *(Vautard et al., 2005)*. Two applications of MetPhoMod model were used for O_3 simulation in the High Tatras Mountain region for the period 12-14 August 2003. In both cases identical meteorological emissions and ground parameters, as well as initial O_3 concentrations were specified as input data. The first application (appointed as interpolation) considered O_3 data from measurement incorporated into the boundary conditions. Results of interpolation can show the agreement of relationship between modelled and measured O_3 concentration. Furthermore the detailed vertical O_3 distribution can clearly illustrate the course of O_3 concentration during the episode. The second simulation calculated O_3 concentrations from local emission sources of model domain and measured O_3 data were not included in the border section. In this case, comparison of modelled and measured O_3 concentration can specify contribution of emissions from local independent sources. Model results for ground level (contains the values of all grid cells just above the ground) are presented.

3.1. Model interpolation of O_3 concentration measured at meteorological observatories and stations of the High Tatras Mountain model domain

The model interpolates O_3 data incorporated as individual O_3 data file into the boundary conditions. The O_3 data file contains O_3 concentrations measured at the stations: Poprad - Gánovce, Stará Lesná, Štart, Skalnaté Pleso and Lomnický štít, adjusted for each vertical layer. Initial O_3 concentrations were identical with measured and other initial background gaseous species concentrations were assigned as zero. Comparison of modelled and measured O_3 concentrations shows small differences (Fig. 6). Correlation coefficients in range from 0.867 to 0.999 also confirm an acceptable accuracy of model interpolation. Sufficient agreement between experimental and modelled O_3 concentration (correlation coefficient r = 0.833) was detected also at station Starý Smokovec. In this case O_3 measurement was excluded from O_3 data file.

Hourly O_3 concentrations obtained by model interpolation were used for illustration of vertical profile of ground level ozone during 12-14 August 2003 in the High Tatras Mountain (Fig. 7). Vertical distribution of O_3 concentrations shows ozone transport from ozone enriched high troposphere to surface layer of atmosphere. Extremely high O_3 concentration ($\sim 190 \mu g\ m^{-3}$) that occurred in the night from 13 to 14 August 2003 on the top layer of model domain (around station Lomnický štít) cannot be caused by photochemical production from local sources. Results of model interpolation support the assumption that O_3 polluted air masses were transported through high tro-

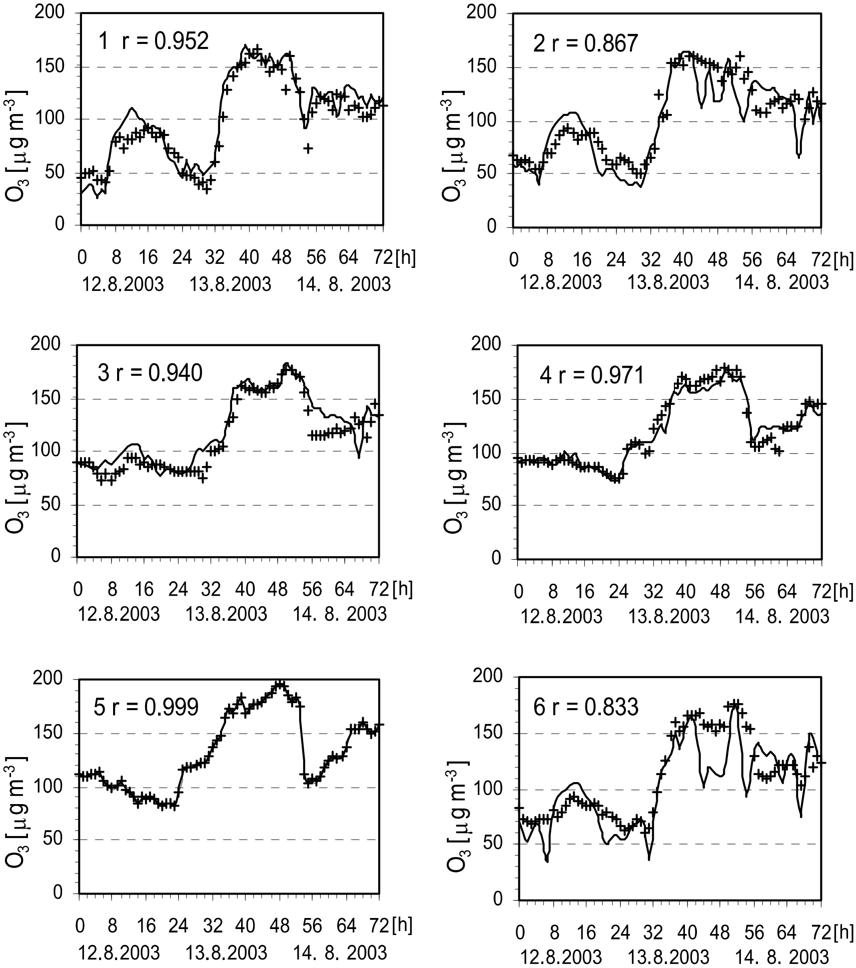

Fig. 6. Hourly O_3 concentrations [μg m^{-3}] of model interpolation (line) and measured data (cross): 1 – Poprad - Gánovce, 2 – Stará Lesná, 3 – Štart, 4 – Skalnaté Pleso, 5 – Lomnický štít, 6 – Starý Smokovec for the period 12-14 August 2003.

posphere from Western Europe to ground layer of atmosphere in the High Tatras Mountain region.

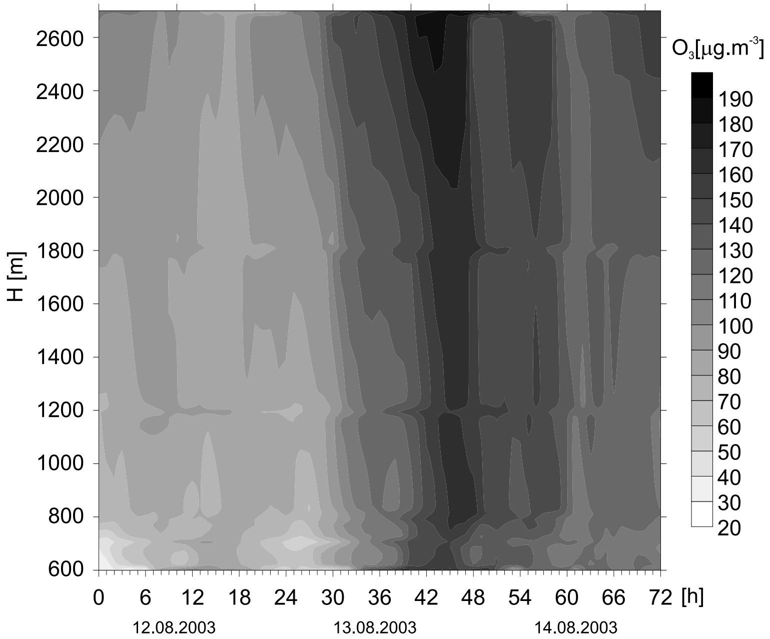

Fig. 7. Vertical profile of hourly O_3 concentrations [μg m^{-3}] obtained by model interpolation for the High Tatras Mountain model domain during 12-14 August 2003.

3.2. Model simulation of O_3 concentration from local emission sources of the High Tatras Mountain domain

The model simulation assumes O_3 concentrations that can be produced by local anthropogenic and biogenic emission sources at the model domain. However, initial O_3 concentrations were defined by measured values, the effect of O_3 measurement on output O_3 values sharply decreased after initialisation, because O_3 data file was excluded from boundary conditions. Another initial gaseous species concentrations were derived from output data file of model interpolation. It is considered that calculating process was not controlled by measured O_3 data and simulated O_3 concentrations represent local ozone forming potential. Daily courses of simulated and measured hourly O_3 concentrations (Fig. 8) show the highest contrast in the night time from 13 to 14 August 2003 (36-54 hours of ozone episode) at all stations of the vertical profile. According to the statistical analysis, the ratio between measured and simulated maximal hourly O_3 concentrations M/S in Table 1 on the first day (August 12) is evidently lower (7-30%) than

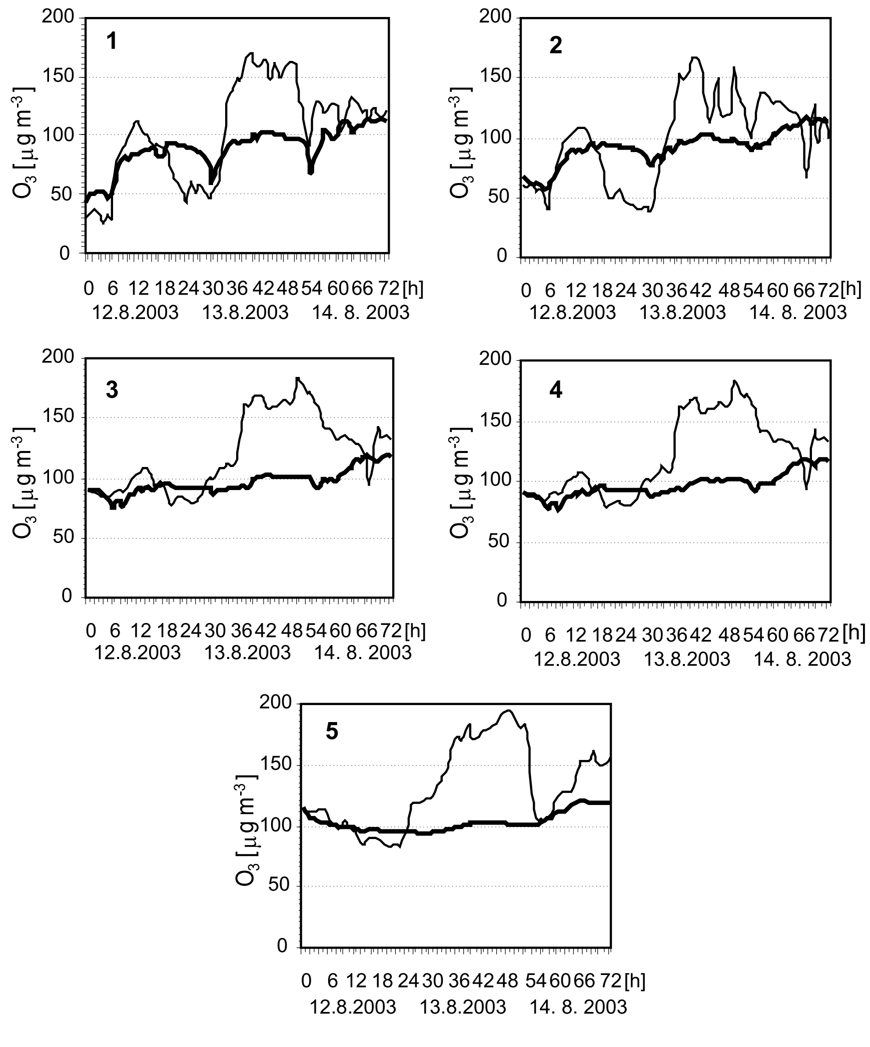

Fig. 8. Hourly O_3 concentrations [μg m^{-3}] measured (thin line) and calculated by model simulation (solid line): 1 – Poprad - Gánovce, 2 – Stará Lesná, 3 – Štart, 4 – Skalnaté Pleso, 5 – Lomnický štít during 12-14 August 2003.

Table 1. Comparison between simulated (S) and measured (M) values of O_3 concentration during 12-14 August 2003 O_3 episode. Maximal hourly and mean daily O_3 concentrations [μg m^{-3}] for individual days and average hourly O_3 concentrations [μg m^{-3}] during ozone episode peak phase (time period from 12 h August 13 to 06 h August 14 2003) are presented separately

Maximal hourly O_3 concentrations [μg m^{-3}]												
Day	12. 08. 2003				13. 08. 2003				14. 08. 2003			
Station	h	M	S	M/S	h	M	S	M/S	h	M	S	M/S
Poprad-Gánovce	12	110.6	84.8	1.30	15	169.9	96.4	1.76	1	161.5	98.1	1.65
Stará Lesná	14	107.0	87.2	1.23	16	165.1	97.6	1.69	2	157.8	98.3	1.61
Štart	13	107.2	88.5	1.21	17	168.3	100.0	1.68	2	182.8	101.0	1.81
Skalnaté Pleso	11	100.3	92.5	1.08	23	164.6	101.7	1.62	0	165.9	101.2	1.64
Lomnický štít	10	105.6	99.0	1.07	23	193.4	101.9	1.90	0	194.8	101.7	1.92

Mean daily O_3 concentrations [μg m^{-3}]									
	M	S	M/S	M	S	M/S	M	S	M/S
Poprad-Gánovce	71.1	75.0	0.95	113.8	90.6	1.22	124.0	101.6	1.23
Stará Lesná	74.3	79.4	0.94	100.5	92.2	1.07	119.7	102.6	1.18
Štart	90.6	87.8	1.04	127.8	94.7	1.34	142.6	105.1	1.37
Skalnaté Pleso	88.7	93.8	0.95	132.1	96.4	1.36	137.9	106.2	1.31
Lomnický štít	96.7	99.6	0.97	153.0	98.3	1.55	145.3	109.4	1.34

Average hourlyO_3 concentrations [μg m^{-3}] during time period from 12 h August 13 to 06 h August 14 2003						
	M	S	M/S	M - S	M – S/M [%]	S/M [%]
Poprad-Gánovce	147.1	96.1	1.53	51.0	34.7	65.3
Stará Lesná	135.1	97.0	1.39	38.1	28.2	71.8
Štart	162.9	99.0	1.65	63.9	39.2	60.8
Skalnaté Pleso	161.0	100.5	1.60	60.5	37.6	62.4
Lomnický štít	177.0	101.5	1.74	75.6	42.7	57.3

on the following days (61-92%) of the ozone episode. Mean daily O_3 concentrations particularly increased at the Lomnický štít. The measured values were substantially higher than the simulated by about: 53% at Poprad - Gánovce, 39% at Stará Lesná, 65% at Štart, 60% at Skalnaté Pleso and 74% at Lomnický štít during peak phase of ozone episode from 12 h (UTC) August 12 to 06 h (UTC) August 13, 2003. In this study, local ozone forming potential is derived from the ratio between the simulated and measured values (S/M in Table 1). According to these results, the contributions to observed O_3 concentration were specified: 63% from local and 37% from local independent (M-S/M in Table 1) emission sources in the High Tatras Mountain region during the peak phase of the ozone episode in August 2003.

Good agreement between MetPhoMod model results and O_3 data obtained by aircraft measurement was achieved during an intensive field campaign in the Grenoble region in July 1999. It is interesting, that the maximum O_3 concentrations were not produced in Grenoble city but at higher altitudes, up to 1500-2000 m a.s.l. over the rural area. Above the residual layer between 1300 and 2300 m a.s.l. O_3 concentration decreased and measured values at 3200 m a.s.l. were considered as ozone background reference level *(Couach et al., 2003)*. It is opposite situation in comparison with the vertical distribution of O_3 concentration in the High Tatras Mountain region in August 2003. Maximal ozone pollution was detected on the top of the model domain at altitude above 2600 m a.s.l. and toward the lower altitudes O_3 concentrations slightly decreased during the peak phase of the O_3 episode.

Assuming relevant contribution of long-range transport, the national reduction of emissions is not an effective tool to achieve decrease of O_3 concentrations in Slovakia. The first results of model LOTOS – EUROS simulation obtained based on the Dutch-Slovak cooperation also show an insignificant impact of Slovak emissions reduction on O_3 concentrations *(Kremler, 2006)*. Furthermore, the increase of O_3 concentrations is expected due to effects of variables associated with future changes in climate and ozone precursor emissions. Climatic changes assumed for temperature, atmospheric water vapor, and biogenic VOC, each individually cause a 1-5% increase in the daily peak ozone in central California *(Steiner et al., 2006)*. Considering unchanged anthropogenic precursor emissions, 10% higher daily maximum values of O_3 concentrations were simulated by coupled climate-chemistry model for the region of southern Germany *(Forkel and Knoche, 2006)*.

4. Conclusion

The model study of O_3 air pollution is a tool for analysing the role of emission sources in extremely high O_3 concentration episode over the High Tatras Mountain region. Coupled model MetPhoMod and measurement were used to investigate the summer ozone episode during 12-14 August 2003. Measured data from fixed ground stations of the model domain (20 km $\times$ 18 km) defined the initial and boundary meteorological conditions for model processing. The comparison between model meteorological input and measured data expressed by correlation coefficient values suggests a good agreement. The spatial distribution of anthropogenic (EMEP database) and BVOC (model GLOBEIS-BEIS2) O_3 precursor concentrations obtained by MetPhoMod corresponds with stationary and mobile emission sources distribution and forested area at the model domain, respectively. Two applications of MetPhoMod model considering O_3 concentrations from local emission sources under meteorological conditions were performed: (1) interpolation - measured O_3 concentrations were involved as individual file into the border conditions; (2) simulation - measured O_3 data were excluded from border section. Vertical profile of O_3 concentration obtained by model interpolation documents ozone transport from ozone enriched high troposphere to surface layer of atmosphere. Extremely high O_3 concentration ($\sim 190 \mu$g m^{-3}) occurred on the top of the model domain (around station Lomnický štít) in the night from 13 to 14 August 2003. Comparison between measured and simulated O_3 concentrations indicates significant differences during the peak phase (from August 12, 12 h UTC to August 13, 06 h UTC). For this time period, mean hourly measured O_3 concentrations were higher by about: 53% at Poprad - Gánovce, 39% at Stará Lesná, 65% - Štart, 60% - Skalnaté Pleso and 74% - Lomnický štít than the simulated ones. Study results suggest that the contribution of transboundary transport to O_3 concentration was on average 37% during peak phase of O_3 episode in August 2003 in the High Tatras Mountain region.

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