

# The EU-25 Emissions from Road Transport and Ground Level Ozone Air Pollution at Rural Mountain Sites in Slovakia

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**Abstract:** Road transport sector produces a significant quantity of ground level ozone ( $O_3$ ) precursors. Relevant abatement of exhaust emissions has been achieved in Europe during last decades due to environmental policy. Presented results indicate relatively weak response of key  $O_3$  precursors decrease:  $NO_x$  (-40%), NMVOC (-69%), and CO (-63%) from the EU-25 road transport to  $O_3$  concentration at background site Stará Lesná in Slovakia for time period 1992-2005. At this site mean annual  $O_3$  concentration varies around  $63.4 \mu\text{g m}^{-3}$  and detected slight decrease of  $-0.89 \mu\text{g m}^{-3}/\text{decade}$  is not statistically significant. Furthermore, high  $O_3$  concentrations occurred in the High Tatra Mts region during the August 2003 heat wave which showed an important impact of long-range air pollution on air quality of Slovak mountain region. More studies will be needed to specify the role of meteorological and environmental factors in the transport and transformation of emissions from large urban regions to remote sites.

**Keywords:** Road transport, emissions, ground level ozone, mountain region,  $O_3$  precursors,  $CO_2$ , GHG.

## INTRODUCTION

Road transport is considered to be an important source of air pollution. The main automobile exhaust components are nitrogen ( $N_2$ ), carbon dioxide ( $CO_2$ ), water vapour ( $H_2O$ ), particulate matter (PM) and reactive ground level ozone ( $O_3$ ) precursors as nitrogen oxides ( $NO_x$ ), non-methane volatile organic compounds (NMVOC), and carbon monoxide (CO). The most widely known photochemical smog is formed in the surface layer of the atmosphere by the reaction between gaseous  $NO_x$  and NMVOC in the presence of strong sunlight. Photochemical events appearing in spring and summer largely consist of high  $O_3$  concentrations. The ozone is a pollutant with potentially toxic effects that can damage human health, trees and plants, and a wide range of natural and artificial materials.

Emissions of  $NO_x$ , NMVOC, CO and PM are regulated for most vehicle types by European emission standards defined in series of the EU directives<sup>1</sup>. European emission standards specify the acceptable limits for exhaust emissions of new vehicles sold in EU member states.

$CO_2$  is an important anthropogenic greenhouse gas (GHG). After power generation, road transport is the second biggest source of greenhouse gas emissions in the EU. It contributes about one-fifth of the EU's total emissions of

$CO_2$ <sup>2</sup>. Currently, there are no standards for  $CO_2$  emissions in the European Union. To help reduce greenhouse gas emissions and meet the Kyoto Protocol targets, the European Commission adopted a proposal COM(2007)856 for legislation to reduce the average  $CO_2$  emissions of new passenger cars which account for about 12% of the EU's carbon emissions. The proposed legislation ensures that average emissions from the new passenger car fleet in the community do not exceed  $120 \text{ g } CO_2/\text{km}$  by 2012. This corresponds to fuel consumption of  $4.5 \text{ l}/100 \text{ km}$  for diesel cars and  $5 \text{ l}/100 \text{ km}$  for petrol cars. Integrated approach including improved engine technology, fuel efficiency, biofuel use, and other specific measures (e.g. penalty premium for car manufacturers) will be used to achieve planned target.

Sensitive  $O_3$  balance is influenced by complex chemistry and synergistic interaction between anthropogenic and biogenic emissions [1] upon varying meteorological and environmental conditions. Emissions produced in large urban regions are transformed into secondary air pollution in atmosphere and through long-range transport affect remote areas. For example, measurement of the National Monitoring network of air quality in Slovakia shows that mean daily  $O_3$  concentration at rural sites is higher than at urban ones during warm weather situations [2]. AOT 40 exposure index that characterizes the harmful effects of  $O_3$  on vegetation is exceeded in a large part of Slovakia in case of forest vegetation and throughout the Slovak territory for agricultural crops [3].

This paper focuses on the association between the emissions from road transport of 25 member states of the European Union (EU-25) and ground level ozone pollution at rural mountain sites in Slovakia.

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<sup>1</sup>EUR-Lex, European Union Law database: Euro 1 (1993): for passenger cars - 91/441/EEC and also for light trucks - 93/59/EEC; Euro 2 (1996) for passenger cars - 94/12/EC (& 96/69/EC); Euro 3 (2000) for any vehicle - 98/69/EC; Euro 4 (2005) for any vehicle - 98/69/EC (& 2002/80/EC), Euro 5 (2008/9) for any vehicle - (COM(2005) 683 - proposed).

<sup>2</sup>EU strategy to reduce  $CO_2$  emissions from cars, <http://europa.eu/>

### NO<sub>x</sub>, NMVOC, CO, AND CO<sub>2</sub> EMISSIONS FROM ROAD TRANSPORT IN THE EU-25, 1992-2005

Expert Emissions data [4] from the UNECE/EMEP<sup>3</sup> emission database WebDab selected for criterions: countries – EU25; years – 1992-2005; pollutants – CO, NO<sub>x</sub>, NMVOC; sector – road transport S7 were used. A detailed description of the methodology can be found in EMEP/CORINAIRE Emission Inventory Guidebook [5]. CO<sub>2</sub> data was obtained from UNFCCC<sup>4</sup> GHG Emissions database selected for EU-25 parties and 1.AA.3.B category of road transport. Fig. (1) clearly shows the decreasing of emissions for reactive chemical components in the EU-25 during 1992-2005 period:

- NO<sub>x</sub> from 7.2 to 4.3 Mt (-40%), with projection 3.3 Mt (2010), 1.8 Mt (2020);
- NMVOC from 5.8 to 1.8 Mt (-69%), with projection 1.2 Mt (2010), 0.8 Mt (2020);
- CO from 32.7 to 12.2 Mt (-63%), with projection 10.4 Mt (2010), 7.8 Mt (2020).

Reduction is in accordance with the strategy to simultaneously remove CO, HC and NO<sub>x</sub> in motor exhaust system using three-way catalyst including:

1. reduction of NO<sub>x</sub> to N<sub>2</sub>:  $2NO_x \rightarrow xO_2 + N_2$
2. oxidation of CO to CO<sub>2</sub>:  $2CO + O_2 \rightarrow 2CO_2$
3. oxidation of unburnt HC to CO<sub>2</sub> and H<sub>2</sub>O:  
 $2C_xH_y + (2x+y/2)O_2 \rightarrow 2xCO_2 + yH_2O$

Reaction scheme (2, 3) demonstrates the conversion of harmful gases (CO, HC) to greenhouse gases CO<sub>2</sub> and H<sub>2</sub>O. UNFCCC data (Fig. 1) documents nearly 21% increase CO<sub>2</sub> emissions from 725 Mt (1992) to 875 Mt (2005) in the EU-25. The absolute quantity of CO<sub>2</sub> in range 700-900 Mt is markedly higher in comparison with amount 2-8 Mt for NO<sub>x</sub>, 1-6 Mt for NMVOC and 5-35 Mt for CO.

Increase of the CO<sub>2</sub> and H<sub>2</sub>O in the atmosphere probably relates to the global warming [6]. Relative contribution of H<sub>2</sub>O and CO<sub>2</sub> to the greenhouse effect is hard to specify, because of their infrared spectrums overlap. Contribution of H<sub>2</sub>O (mostly as water vapour, and as clouds) to the GHG effect is a significantly more than CO<sub>2</sub>. The contribution of clouds is important, greater than that of CO<sub>2</sub>, but less than that of water vapour in the atmosphere [7]. The amount of warming depends on various feedback mechanisms. Due to rising levels of GHG concentration, water vapour increases and causes more warming, this causes an additional increasing of water vapour, in a self-reinforcing cycle. This water vapour feedback may be strong enough to approximately double the increase in the greenhouse effect due to the added CO<sub>2</sub> alone [6].

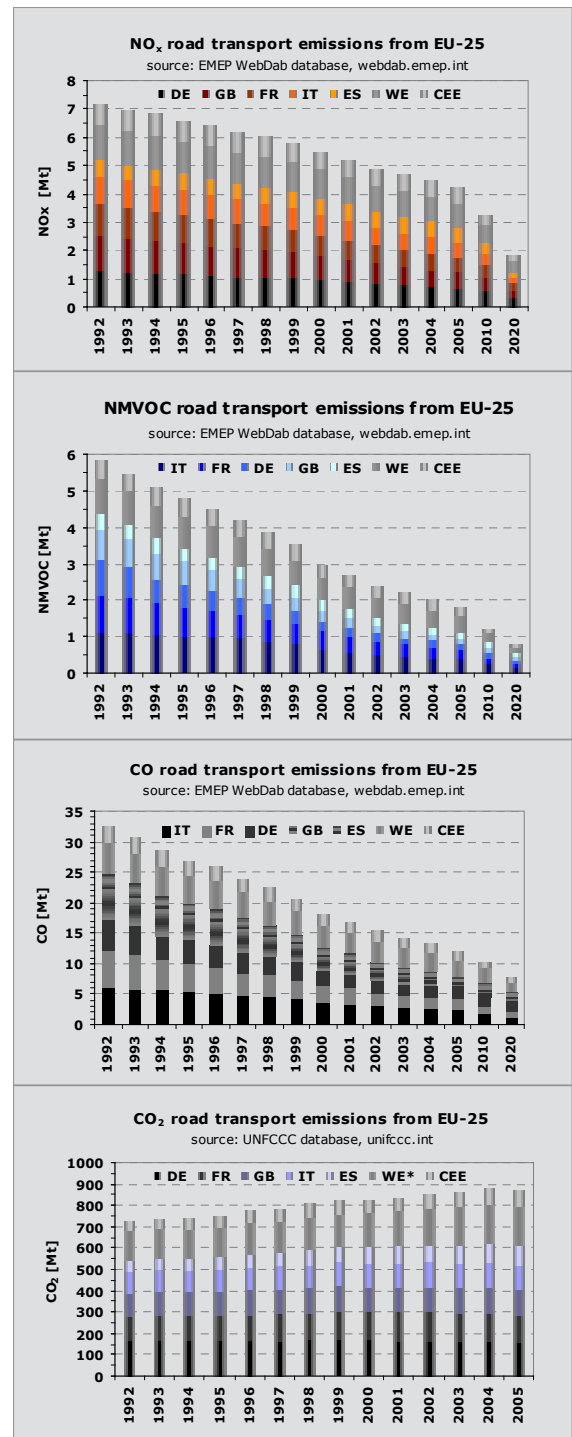


Fig. (1). Road transport emissions: O<sub>3</sub> precursors – NO<sub>x</sub>, NMVOC, CO, and GHG – CO<sub>2</sub> produced by the EU-25 countries<sup>5</sup> in 1992 – 2005.

<sup>3</sup> EMEP is a scientifically based and policy driven program under the Convention on Long-range Transboundary Air Pollution for international co-operation to solve transboundary air pollution problems.

<sup>4</sup> UNFCCC - United Nations Framework Convention on Climate Change ([www://unfccc.int/2860.php](http://www.unfccc.int/2860.php)) is an international environmental treaty produced at the United Nations Conference on Environment and Development (UNCED), informally known as the Earth Summit, held in Rio de Janeiro in 1992. The treaty is aimed at reducing emissions of greenhouse gas in order to combat global warming.

<sup>5</sup> Main producers: DE-Germany, FR-France, GB-United Kingdom, IT-Italy, ES-Spain; other WE - West Europe countries: AT-Austria, BE-Belgium, CY-Cyprus, DK-Denmark, FI-Finland, GR-Greece, IE-Ireland, LU-Luxembourg, MT-Malta, NL-Netherlands, PT-Portugal, SE-Sweden \*Malta and Cyprus did not provide GHG emission estimates, therefore CO<sub>2</sub> data are not included to WE group; CEE - Central and East Europe countries: CZ-Czech Republic, EE-Estonia, H-Hungary, LV-Latvia, LT-Lithuania, PL-Poland, SK-Slovakia, SI-Slovenia.

**GROUND LEVEL OZONE IN THE HIGH TATRA Mts**

Ground level ozone is a serious problem in the High Tatra Mts region (HTMts). Air pollution has a negative impact on sensitive ecosystems of the Tatra National Park (TANAP) biosphere reserve [8]. Monitoring of O<sub>3</sub> concentration and the standard meteorological measurements are provided at stations situated in altitude profile from 706 to 2634 m a.s.l. (Fig. 2) by the Slovak Hydrometeorological Institute (SHMI) in cooperation with the GPI SAS and the Research Centre of the Tatra National Park (RC TANAP). O<sub>3</sub> concentration is measured by UV absorption ozone analysers. Equipments are adjusted in accordance with the secondary national ozone calibration standard of SMHI and intercomparisons with the Czech primary ozone standard are regularly organized.



site	H (m a.s.l.)	Lat (φ)	Long (λ)
Lomnický štít	2634	49°12' N	20°13' E
Skalnaté Pleso	1778	49°11' N	20°14' E
Štart	1200	49°10' N	20°15' E
Stará Lesná	810	49°09' N	20°17' E
Poprad-Gánovce	706	49°02' N	20°19' E

Fig. (2). Altitude profile of ground based measurements for O<sub>3</sub> concentration and standard meteorological parameters in the High Tatra Mts region.

**Road Emissions in the EU-25 and O<sub>3</sub> Concentration at the Station Stará Lesná**

The station Stará Lesná is situated in a locality without large mobile and stationary emission sources. Therefore, observed O<sub>3</sub> concentrations are affected mainly by wide regional and long - range transport of air pollution in relation to weather and wind situation. The High Tatras Mts represent a topographical barrier, especially for the north components of general atmospheric circulation. The prevailing airflow components are the S and SSW at Stará Lesná and WSW and SW at Skalnaté Pleso [9]. Air pollution from South and Western Europe including transformed road transport emissions can also influence the O<sub>3</sub> level in this region. Fig. (3) presents the simultaneous course of annual emissions from road transport of the EU-25 countries and annual means of O<sub>3</sub> concentration at Stará Lesná for time period 1992-2005. Data obtained at the MO Stará Lesná represent the longest continuous time series of O<sub>3</sub> concentration for rural sites in Slovakia. Lines of Fig. (3) indicate that an evident decrease of NO<sub>x</sub>, NMVOC and CO does not correspond to variable O<sub>3</sub> concentration. Average annual O<sub>3</sub> concentration varies around mean value of 63.4 μg m<sup>-3</sup> (1992-2005) in range from 52.1 μg m<sup>-3</sup> (1998) to 71.8 μg m<sup>-3</sup> (1996) and detected slight decrease of -0.89 μg m<sup>-3</sup>/decade

is not statistically significant. Statistical analysis shows that the information threshold<sup>6</sup> 180 μg m<sup>-3</sup> was exceeded more often in 1992 (7 times) than in 1999 (twice) and alert threshold 240 μg m<sup>-3</sup> was not overstepped during the whole considered period. Maximal daily mean 150 μg m<sup>-3</sup> in April 4, 1996 and hourly maximum 226 μg m<sup>-3</sup> in March 11, 1999 at 13 h were recorded.

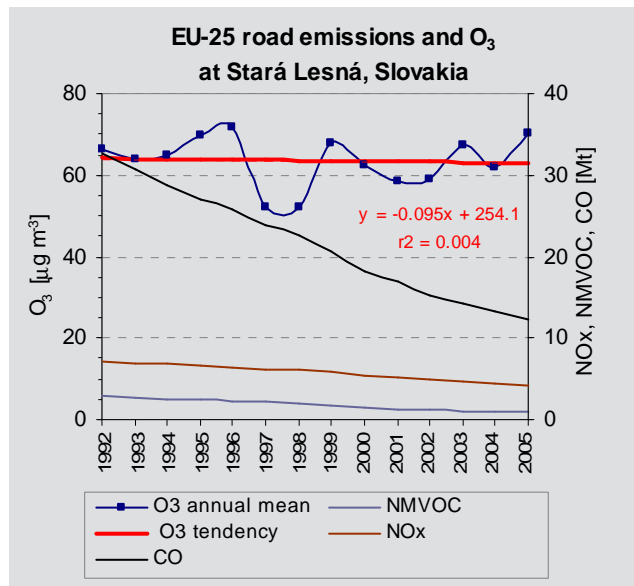


Fig. (3). EU-25 road emissions and course of annual mean O<sub>3</sub> concentration [μg m<sup>-3</sup>] with a tendency for rural station Stará Lesná in 1992-2005.

There are two maxima in annual course - the primary in March-April and the secondary in August. The primary maximum is related to convenient meteorological conditions after the end of winter period. Abundance of reservoir O<sub>3</sub> precursors (NO<sub>3</sub>, HNO<sub>3</sub>), increase of solar irradiance and air temperature, and decrease of relative air humidity provide appropriate conditions for progressive O<sub>3</sub> photochemical production in spring [10]. Biogenic volatile organic compounds (BVOC) [11] and secondary organic aerosol (SOA) probably play a serious role in summer O<sub>3</sub> formation. This suggestion is supported by findings of EMEP EC/OC campaign [12] that show higher contribution of organic carbon (OC) than elemental carbon (EC) for summer than winter at Stará Lesná. Especially high O<sub>3</sub> concentrations associated with the long-range transport of air pollution occur during extreme weather events in summer e.g. in August 2003.

**O<sub>3</sub> Pollution in the High Tatra Mts During Extreme Weather Situation**

Exceptional 15-day long heat wave with record temperatures and unusually persistent high-ozone concentrations was observed in Europe in August 2003 [13]. Monitoring stations in Slovakia also recorded high maximal O<sub>3</sub> concentrations in the range of 127 to 301 μg m<sup>-3</sup>. The ambient air quality standards were exceeded frequently in Bratislava and at Lomnický štít: alert threshold 20 times only in Bratislava, information threshold total 69 times. Increase of O<sub>3</sub> concentration with altitude documents measurement in HTMts vertical profile. Mean monthly values raised from 82 μg m<sup>-3</sup> at

<sup>6</sup> Directive 2002/3/EC of the European Parliament and of the Council of 12 February 2002 relating to ozone in ambient air.

Stará Lesná to  $124 \mu\text{g m}^{-3}$  at Lomnický štít in August 2003 [2].

The summer  $\text{O}_3$  episode observed during August 12-14, 2003 in HTMTs region was investigated by model MetPhoMod [14]. Model applications [15] included: 1 - interpolation of measured  $\text{O}_3$  data, and 2 - simulation of  $\text{O}_3$  concentrations produced mainly from local emission sources. Hourly  $\text{O}_3$  data obtained by model interpolation were used for illustration of  $\text{O}_3$  vertical profile (Fig. 4). Fig. (4) shows ozone transport from higher tropospheric layer to surface layer of atmosphere. The highest  $\text{O}_3$  concentration ( $\sim 190 \mu\text{g m}^{-3}$ ) occurred in the night from 13th to 14th August 2003 in the altitude above 2600 m a.s.l. can not be caused by photochemical production from local sources. Results of model interpolation correspond with the ozone forecast provided by the French information system PREV'AIR (Fig. 5) that indicates transport of  $\text{O}_3$  polluted air masses from Western Europe to the east after August 12, 2003. Daily course of simulated and measured hourly  $\text{O}_3$  concentrations (Fig. 6) shows the highest contrast in the night time from 13 to 14 August 2003 at the station Lomnický štít.

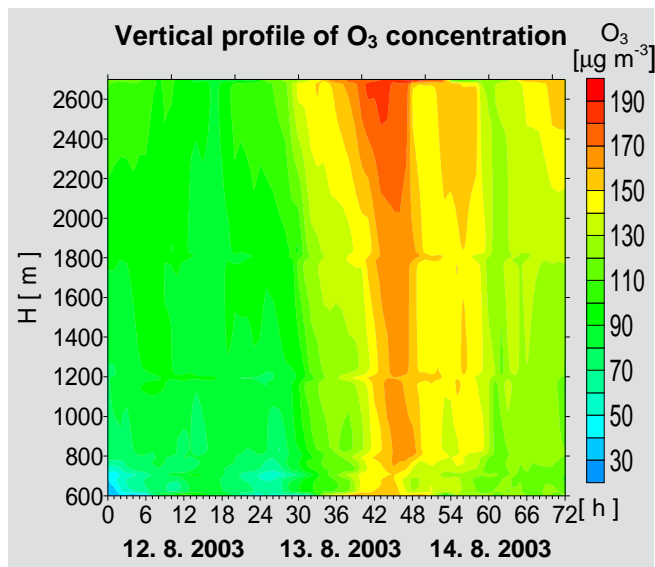


Fig. (4). Vertical profile of  $\text{O}_3$  concentrations [ $\mu\text{g m}^{-3}$ ] obtained by MetPhoMod model interpolation in the High Tatra Mts in August 12-14, 2003.

Comparative analysis of measured (M), simulated (S), and complementary (C)  $\text{O}_3$  concentration (Fig. 7) was used for specifying the contribution of local independent sources to observed  $\text{O}_3$  concentrations during peak phase of the ozone episode from 12 h (UTC) August 12 to 06 h (UTC) August 13, 2003.

Obtained results (Fig. 8):

- measured  $\text{O}_3$  concentration raised in range from  $135 \mu\text{g.m}^{-3}$  (Stará Lesná) to  $177 \mu\text{g.m}^{-3}$  (Lomnický štít) were substantially higher than simulated about: 53% at Poprad-Gánovce, 39% at Stará Lesná, 65% at Štart, 60% at Skalnaté Pleso and 74% at Lomnický štít;
- the highest complementary  $\text{O}_3$  concentration was  $76 \mu\text{g.m}^{-3}$  - it represents 43% contribution to measured  $\text{O}_3$  concentration at Lomnický štít indicate serious influence of local independent emission sources to maximal  $\text{O}_3$  concentrations

in the High Tatra Mts region during the extremely weather situation in August 2003.

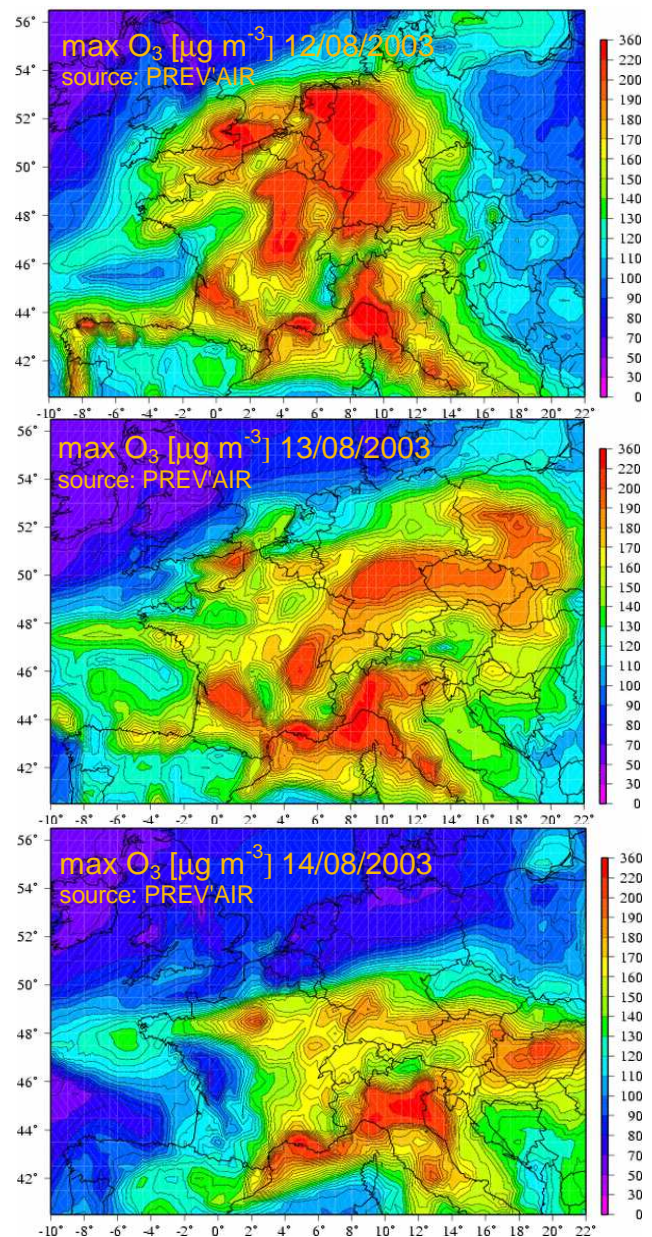


Fig. (5). PREV'AIR forecast <http://www.prevail.org/en/index.php> of daily peak of  $\text{O}_3$  concentrations for the time period August 12-14, 2003.

Assuming relevant contribution of air pollution from long-range transport, the national reduction of emissions is not an effective tool to achieve decrease in  $\text{O}_3$  concentrations in Slovakia. The first results of model LOTOS – EUROS simulation also show an insignificant impact of Slovak emissions reduction to  $\text{O}_3$  concentrations [16]. Furthermore, increase of  $\text{O}_3$  level is expected due to effects of variables associated with future changes in climate and  $\text{O}_3$  precursor emissions. Considering unchanged anthropogenic precursor emissions, 10% higher daily maximum values of  $\text{O}_3$  concentrations were simulated by coupled climate-chemistry model for the region of southern Germany [17]. Climatic changes assumed for temperature, atmospheric water vapour, and

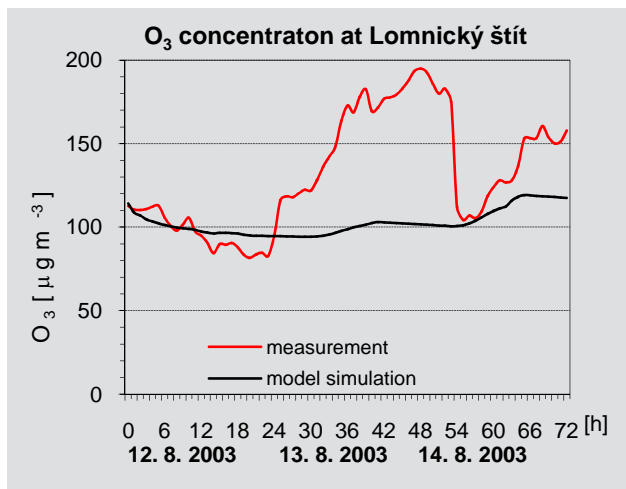


Fig. (6). Comparison of measured (red) and simulated (black) O<sub>3</sub> concentrations [µg m<sup>-3</sup>] at the station Lomnický štít in August 12-14, 2003.

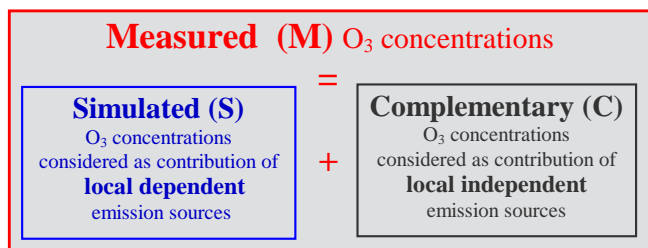


Fig. (7). Scheme of relationship among measured (M), simulated (S), and complementary (C) O<sub>3</sub> concentrations.

BVOC, each individually causing a 1-5% increase in the daily peak ozone in central California [18]. Measurements and modeling results suggest that there is a strong link between climate change and surface ozone. A warmer and dryer European climate is very likely to increased O<sub>3</sub> concentrations. Furthermore, increased anthropogenic emissions in developing economies in Asia are likely to raise the hemispheric background level of ozone. Gradually, these effects may outweigh the effect of the reduced European ozone precursor emissions. This calls for a global or hemispheric perspective in the revision of the European air quality policy for ozone [19].

**CONCLUSIONS**

The control of air pollution is essential for sustainable development of society. Environmental policy of the European Union contributes to relevant reduction of emissions from road transport in Europe. Abatement has been achieved for key O<sub>3</sub> precursors: NO<sub>x</sub> (-40%), NMVOC (-69%), and CO (-63%) produced by vehicles in the EU-25 during 1992-2005. On the other hand, measurement at the background station Stará Lesná in Slovakia indicates weak response of emission reduction on O<sub>3</sub> concentration. At this site average annual O<sub>3</sub> concentration varies around mean value of 63.4 µg m<sup>-3</sup> (1992-2005) in range from 52.1 µg m<sup>-3</sup> (1998) to 71.8 µg m<sup>-3</sup> (1996) and detected slight decrease of -0.89 µg m<sup>-3</sup>/decade is not statistically significant. Besides this, extraordinary O<sub>3</sub> event in the summer 2003 underlines serious

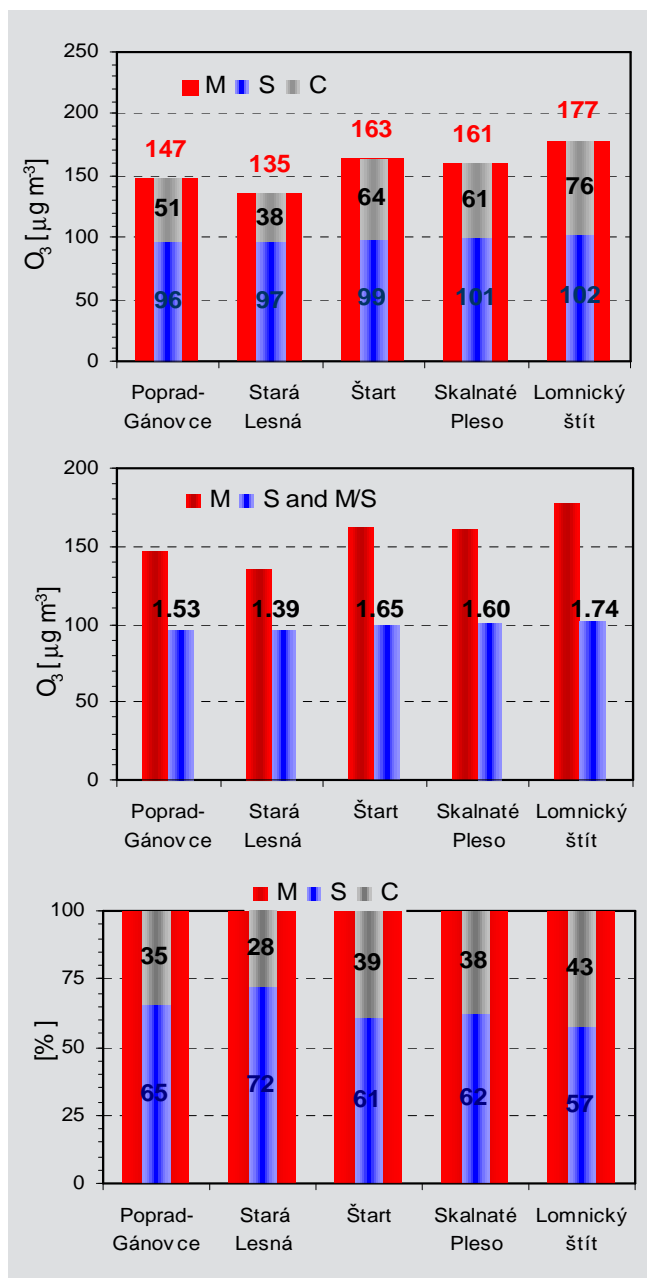


Fig. (8). Mean hourly O<sub>3</sub> concentrations [µg m<sup>-3</sup>]: M - measured, S - simulated, C - complementary for the peak phase of O<sub>3</sub> episode in the High Tatra Mts in August 2003.

problem of transboundary air pollution. Approximately 30-40% contribution of local independent emission sources to measured O<sub>3</sub> concentration was estimated by model simulation for the High Tatra Mts region during peak phase of O<sub>3</sub> episode in August 2003. Vertical profile of O<sub>3</sub> concentration obtained by model MetPhoMod interpolation documents O<sub>3</sub> transport from O<sub>3</sub> enriched high troposphere to surface layer of atmosphere in the heat-wave conditions. Special meteorological situation supports the progressive O<sub>3</sub> accumulation and then its transport through free troposphere for long distances from the original emission sources. The complex approach that involves effects of synergic interactions of emission, meteorological and environmental factors in context of global warming can be useful for modification of air quality strategies.

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