Summer ground level ozone maximum in Slovakia in 2003

S. Bičárová
Geophysical Institute of the Slovak Academy of Sciences[1]

M. Sojáková, C. Burda
Slovak Hydrometeorological Institute[2]

P. Fleischer
Research Centre of the Tatra National Park[3]

Abstract: Extraordinary high level of ozone pollution occurred in Slovakia in August 2003. Monitoring stations recorded the following maximal O_3 concentrations: mean hourly values of 127 to 301 μg m^{-3}, mean daily values of 72 to 153 μg m^{-3}, mean monthly values of 47 to 124 μg m^{-3}. The ambient air quality standards were frequently exceeded at the SW lowlanded urban area in Bratislava and the mountain rural station Lomnický štít: alert threshold IH_{1h} = 240 μg m^{-3} – 6 times in Bratislava, information threshold IH_{1h} = 180 μg m^{-3} – 44 times in Bratislava, 8 times at Lomnický štít, total 69 times at all monitoring stations. The target value 8h mean IH_{8h} = 120 μ g m^{-3} was exceeded also in Bratislava and Lomnický štít over more than 25 days. General comparison of mean daily O_3 concentration of urban and rural stations shows that ozone level at the rural sites is higher than at the urban ones. The summer ozone maximum in August 2003 appears to be associated with special ozone production and distribution due to the unusual warm weather situation and the transport of abundant ozone precursors over Europe. The decreasing trend of NO_x pollutant suggests the ozone and precursors transboundary transport into Slovakia.

Key words: ground level ozone concentration, summer maximum, urban and rural sites, exceedance of the ambient air quality standards

[1] Stará Lesná, 059 60 Tatranská Lomnica, Slovak Republic; e-mail: bicarova@ta3.sk; Web: http://www.ta3.sk/gfu

[2] Jeséniova 17, 833 15 Bratislava 37, Slovak Republic
e-mail: michaela.sojakova@shmu.sk, cyril.burda@shmu.sk; Web: http://www.shmu.sk

[3] 059 60 Tatranská Lomnica, Slovak Republic; e-mail: fleischer@vstanap.sk; Web: http://www.vstanap.sk

1. Introduction

Exceptionally extreme ozone pollution was achieved in Europe in summer 2003. Nevertheless, the emissions of ozone precursors have declined substantially in the last 15 years (*Vestreng et al., 2004; Spišáková et al., 2003*), the measurements in 2003 have shown the highest values at the most of European ground level ozone monitoring stations including Slovakia. On average, exceedance of the 240 μg m^{-3} alert threshold defined in the new Ozone *Directive 2002/3/EC of the European Parliament* was observed in summer 2003 at 27% of 1805 European stations. The maximum mean hourly ozone concentration reported in 2003 was 417 μg m^{-3} at the monitoring station in France *(Fiala et al., 2003).* In Slovakia the alert threshold 240 μg m^{-3} was exceeded only on SW Slovakia in six cases and the mean hourly ozone concentration over 300 μg m^{-3} was observed only in one case at the Bratislava-Petržalka station. The target value 8h mean $\mathrm{IH_{8h}} = 120$ μg m^{-3} was exceeded at most of stations over more than 25 days in 2003 *(Hrouzková et al., 2004).*

Special meteorological situation favoured the progressive accumulation of ozone in August 2003. Marked atmospheric stability with very weak winds, which limited the dispersion of pollutants, the high temperatures caused by the advection of a warm African air mass and strong solar radiation with clear skies led to high ozone concentrations until the disruption of the aforementioned stable atmospheric conditions occurred (*Rodríguez et al., 2004*). No significantly negative trend for daily ozone maxima in Switzerland suggests that the emission reductions of primary pollutants during the 1990s had little impact on the daily maximum ozone concentrations *(Ordonez, 2004).* Ozone precursors can be transported over distances of hundreds to thousands kilometres, resulting in ozone formation far from the sources. Hemispheric background accounts annually for 64.4% of total O_3 burden, while North American, Asian and European contributions due to anthropogenic sources account for 10.9%, 7.7%, and 9.4%, respectively. In the free troposphere, the change in O_3 is driven by the increase in Asian contribution, which compensates the decrease due to the local emission reductions in all seasons *(Auvray* and*Bey, 2004).* Reinforces of international treaties regulating the emissions is needed, because the effect of the European emissions reductions may be offset by the increase of foreign emissions *(Holloway* *et al., 2003).*

The purpose of this paper is to evaluate the summer maximum of extreme high ground level ozone concentration occurred in Slovakia in August 2003 and to provide analysis relative to the environment type of ozone monitoring locality.

2. Materials and methods

Mean hourly ozone concentrations from 21 sites of the Slovak Hydrometeorological Institute (SHMI) monitoring network, 5 stations owned by of the National Park High Tatras Administration during the period from 1 to 31 August 2003 were used in this study. Data are collected and validated in central SHMI database. At all stations, ozone was measured with the UV absorption method. Ozone analyzators are adjusted in accordance with the secondary national ozone calibration standard of SHMI. Intercomparisons with the Czech primary ozone standard are regularly organized.

Directive 2002/3/EC of the European Parliament establishes long-term objectives for concentrations of ozone in ambient air. Target value for the protection of human health: maximum daily 8h mean $IH_{8h} = 120\ \mu g\ m^{-3}$ not to be exceeded on more than 25 days per calendar year averaged over three years. Thresholds values available to the public: mean hourly $IH_{1h} = 180\ \mu g\ m^{-3}$ for information and mean hourly $IH_{1h} = 240\ \mu g\ m^{-3}$ for alert.

Distribution of ground level ozone monitoring stations grouped into categories according to their spatial distribution and land cover use *(Anderson et al., 1976)* is included in Tab. 1 and Fig. 1. Distinguish emission quantity produce mobile and stationary sources particularly in urban agglomeration due to intensive industry, transportation, commercial, services activity and extensive built-up area. Following processes of photochemical transformation, horizontal advection, vertical flow and dispersion in troposphere cause precursors and ozone transport over large surrounding area to rural sites. Rural ozone stations are located in small settlements or areas with natural ecosystems, forests or crops. Majority of rural stations are situated in highland and mountain environment in Slovakia.

Methods of mathematical statistics *(Anděl, 1985; Montgomery and Runger, 1999; Nosek, 1972)* were used for evaluation of described experimental data with focus on comparison and relationship analysis.

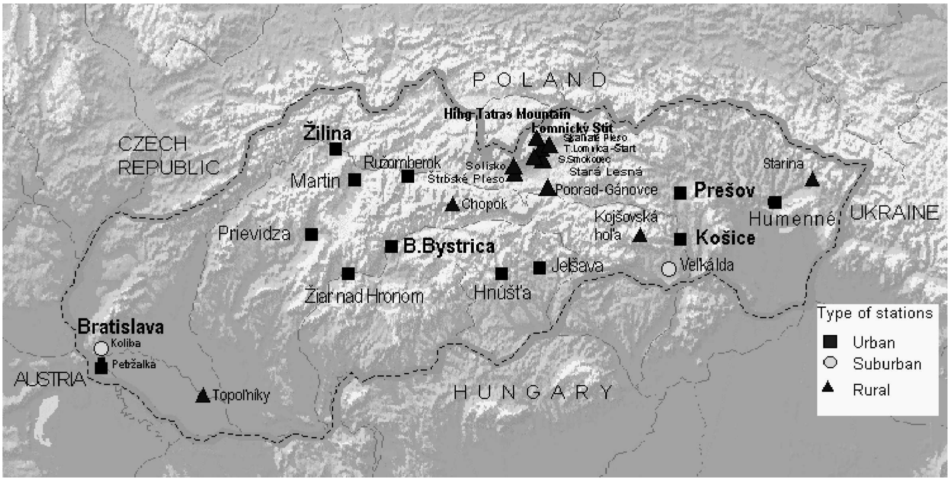

Fig. 1. Monitoring network of ground level ozone stations in Slovakia.

3. Results and discussion

3.1. Analysis of the meteorological situation

Unusual extreme high temperatures in the May-August season and very low precipitation totals in the February-August season were recorded in Slovakia in 2003 (*Faško et al., 2003*). Particularly in August 2003 at the most territory of Slovakia exceedance of long-time values was observed: mean monthly temperature with positive deviations of 2.3° C to 4.8° C; mean relative air humidity at 14 h within –18% to –11%; precipitation deficit 45 mm; sum of sunshine duration of 141% to 205%. The temperature extremes mainly in SW part and the highest temperature maximum 38.8° C in Bratislava on 13 August were achieved. Comparison of wind speed and direction at meteorological stations in Bratislava-Koliba and Lomnický štít in August 2003 is given in Fig. 2. NW-W and NE respectively N and NW prevailing frequency of wind directions at lowland suburban Bratislava-Koliba respectively at mountain Lomnický štít were recorded. Mean wind speed around 0.1 m s^{-1} flowed from all directions and maximal wind speed approximately 0.8 m s^{-1} in Bratislava-Koliba indicate weak dispersion of pollutant. More favourable wind for pollutant transport from SW and W

Tab. 1. List of ground level ozone monitoring stations in Slovakia rearranged into groups: urban and suburban stations, rural stations; within groups sort of stations after descending order of NO_x emission percentage

Station: name, locality	[4]Emission NO_x [%]	Type	Area	[5]Altitude [m a.s.l.]	Latitude N	Longitude E	Evidence data from
URBAN (U) and SUBURBAN (SU) STATIONS							
Košice locality	21.0						
Košice-Veľká Ida		SU	Industrial	207	48°35'	21°10'	1992
Košice-Podhradová		U	Built-up	248	48°45'	21°14'	1996
Prievidza	10.5	U	Services and transport	265	48°46'	18°37'	1992
Bratislava locality	9.2						
Bratislava-Petržalka		U	Built-up and transport	136	48°07'	17°08'	1992
Bratislava-Koliba		SU	Residential	287	48°10'	17°07'	1996
Ružomberok	2.2	U	Built-up and industry	464	49°04'	19°18'	1999
Jelšava	2.1	U	Built-up and industrial	255	48°37'	20°14'	1997
Žilina	1.7	U	Built-up	362	49°12'	18°46'	1994
Žiar nad Hronom	1.4	U	Built-up and industrial	263	48°35'	18°51'	1992
Martin	0.9	U	Transportation	383	49°04'	18°55'	1994
Prešov	0.7	U	Transportation	239	48°59'	21°15'	1998
Banská Bystrica	0.6	U	Transportation	343	48°44'	19°09'	1993
Hnúšťa	0.5	U	Built-up and industrial	315	48°35'	19°57'	1994
Humenné	0.2	U	Commercial and	160	48°55'	21°54'	1994
RURAL STATIONS: Lowland (L), Uplands (Up), Highlands (H), Mountain (M)							
Topoľníky	1.6	L	Natural agriculture	113	47°57'	17°51'	1994
Kojšovská hoľa	1.2	H	Natural forest and	1244	48°47'	20°59'	1999
Starina	0.5	Up	Natural water	345	49°02'	22°15'	1995
Chopok	0.5	M	Natural alpine	2008	48°56'	19°36'	1991
The High Tatras Mountain vertical profile							
Poprad locality	0.4						
Poprad-Gánovce		Up	Natural meadow	706	49°02'	20°19'	1999
Stará Lesná		Up	Natural meadow and	814	49°09'	20°17'	1991
Starý Smokovec		Up	Services and transport	1000	49°08'	20°13'	2003
T. Lomnica - Štart		H	Natural forest	1200	49°10'	20°15'	2000
Štrbské Pleso		H	Natural forest	1361	49°07'	20°03'	2000
Skalnaté Pleso		M	Natural subalpine	1770	49°11'	20°14'	2000
Solisko		M	Natural subalpine	1840	49°08'	20°02'	2003
Lomnický štít		M	Natural apline	2635	49°12'	20°13'	2001

[4] Proportion of local to total stationary source NO_x emissions in Slovakia in 2003 *(Sajtáková, 2004)*.

[5] http://cms.shmu.sk/net.html

is indicated by results recorded at Lomnický štít: mean wind speed approximately 0.8 m s^{-1} and maximal mean hourly wind speed 24.7 m s^{-1}.

3.2. Summer 2003 ozone episode

In general the summer ground level ozone maximum in July or August at the Slovak stations is subordinate. The main maximum is in April *(Krem-*

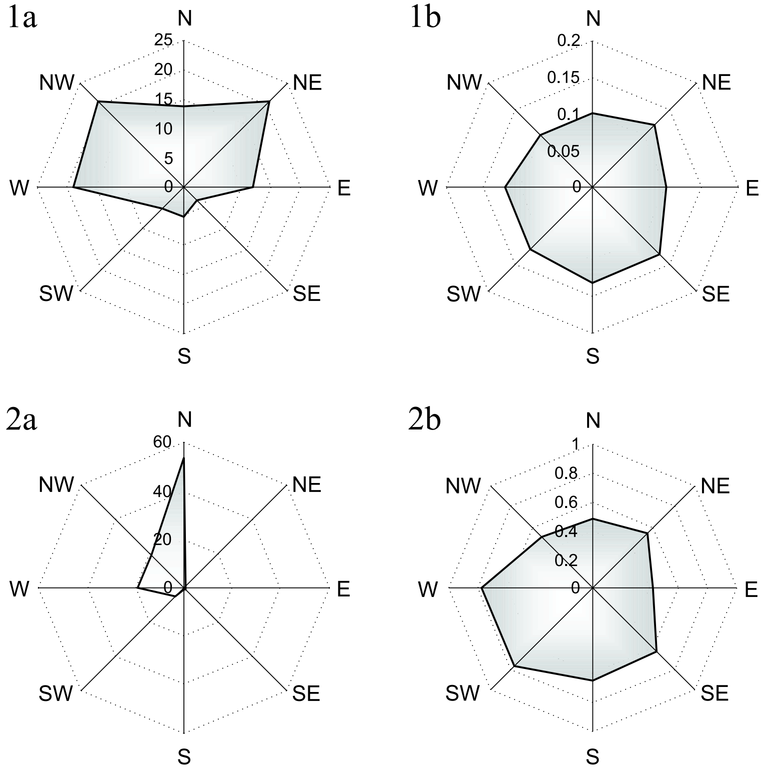

Fig. 2. Wind roses: a - frequency of direction, b - wind speed [m s^{-1}] at meteorological stations: 1 – Bratislava-Koliba, 2 – Lomnický štít.

ler, 2002). Ozone photochemistry process in abundance of NO_x and VOC precursors leading to the highest O_3 concentrations episode is supported by anticyclone circulation. The evaluation of synoptic situation shows alternation of relative long-term anticyclone occasions with weak cold fronts in Slovakia in August 2003. The first progressive advance of high pressure air from NE elevated on 13 August. Extraordinary mean hourly O_3 concentrations in the range from 127 to 301 μg m^{-3} were recorded nearly at all stations on 12–14 August (Tab. 2). The exceptions noted at SE sites, where probably influence of front margins moved from Northern Europe occurred. Other high pressure system developing in second half of August can be associated with maximum mean hourly O_3 concentrations occurred at

NW and E Slovakia on 23 August. The heatwave moving from SW before the end of August appears to influence ozone concentration increase over south of Central Slovakia, because maximal mean hourly O_3 concentration 174 μg m^{-3} in Hnúšťa was reached on 29 August. Mean daily O_3 concentration at all stations achieved a maximum of 73 to 153 μg m^{-3} also during anticyclone progression. The highest ozone level of reached at the mountain site Lomnický štít and at lowlanded stations in Bratislava. Extreme high O_3 concentration corresponds with frequency of excess over the ambient air quality standard: alert threshold $IH_{1h} = 240$ μg m^{-3} 6 times only at ozone stations in Bratislava, information threshold $IH_{1h} = 180$ μg m^{-3} 44 times in Bratislava, 8 times at Lomnický štít, total 69 times at all monitoring stations were exceeded. The number of days when target was value $IH_{8h} =$ 120 μg m^{-3} for the protection of human health was overstepped accounted more than 25 days also in Bratislava and Lomnický štít. Additionaly, at Lomnický štít total number of days with exceedance of target value IH_{8h} achieved 198 days in 2003.

Comparison of mean daily O_3 concentration at urban and rural stations shows (Fig. 3) that ozone level at rural stations is higher than at urban stations. Similar results were presented by *Duenas et al. (2004)* and *Coyle et al. (2002).* In urban areas, O_3 concentrations may be lower than the rural concentrations due to chemical feeding by local NO. In the suburbs and further downwind of large cities, where local NO_x emissions are lower, the formation generally dominates over depletion and elevated O_3 levels are found *(Louka et al., 2003).* At mountain tops reaching into the atmospheric mixing layer, the absence of local NO_x emissions and the minor importance of deposition to the ground as ozone-removing mechanisms lead to less ozone destruction. On the other hand the high emission density of reactive precursors in urban areas might lead to high O_3 values within the city or at short distances as indicate Fig. 3. Positive deviations for urban stations represent maximal mean hourly value and achieved level 300 μg m^{-3} in August 2003. Variability of amplitude - range of minimal and maximal mean hourly O_3 concentrations - at urban stations was substantially higher due to extreme O_3 concentration than at rural stations with higher level of minimal values.

Different variability appears to be associated with differences in ozone daily pattern. The daily course of mean hourly O_3 concentrations depends on the type of station and altitude (Fig. 4). At urban and upland rural

Tab. 2. Statistical characteristics of ground level ozone concentration [μg m^{-3}] in Slovakia in August 2003 rearranged into groups: urban and suburban stations, rural stations; within groups sort of stations after descending order of mean monthly O_3 concentration; the High Tatras Mountain stations sorted after ascending order of altitude

Station	Mean ground level ozone concentrations [μg m^{-3}] in August 2003					Frequency of excess over the ambient air quality standard in August 2003			
	Monthly	Daily		Mean hourly		Thresholds values IH_{1h}		Target value IH_{8h}	
		max	date	Max	date h	180 μg m^{-3}	240 μg m^{-3}	120 μg m^{-3}	Total 2003
URBAN and SUBURBAN									
Bratislava-Koliba	118	148	13.08.	255	12.08. 11:00	27	3	26	78
Košice-Podhradová	93	126	23.08.	178	23.08. 11:00	0	0	18	64
Jelšava	88	113	14.08. 23.08.	169	23.08. 17:00	0	0	20	66
Bratislava-Petržalka	88	120	23.08.	301	12.08. 10:00	17	3	20	55
Humenné	85	107	14.,18., 23.,29.08.	179	23.08. 15:00	0	0	21	68
Hnúšťa	85	113	23.08.	174	29.08. 13:00	0	0	22	79
Žiar nad Hronom	79	98	23.08.	172	14.08. 15:00	0	0	20	66
Prešov	76	110	23.08.	186	23.08. 14:00, 15:00	2	0	21	61
Prievidza	71	110	29.08.	159	13.08. 16:00	0	0	13	33
Banská Bystrica	71	91	23.08.	172	23.08. 13:00	0	0	20	48
Žilina	70	97	29.08.	172	13.08. 15:00 23.08. 12:00,	0	0	20	57
Martin	67	83	23.08.	167	13.08. 16:00	0	0	16	29
Ružomberok	54	72	23.08.	126	23.08. 11:00	0	0	1	6
Košice-Veľká Ida	47	73	23.08.	127	14.08. 13:00, 14:00	0	0	5	-
RURAL									
Kojšovská hoľa	121	148	14.08., 18.08.	174	14.08. 00:00	0	0	21	97
Chopok	120	150	13.08.	184	13.08. 21:00	3	0	24	103
Starina	94	122	14.08., 23.08.	177	14.08. 11:00	0	0	23	67
Topoľníky	92	127	23.08.	232	13.08. 16:00	8	0	26	103
The High Tatras Mountain vertical profile									
Poprad-Gánovce	89	124	14.08.	170	13.08. 15:00	0	0	18	53
Stará Lesná	82	121	14.08.	166	23.08. 14:00	0	0	15	39
Starý Smokovec	89	130	29.08.	176	14.08. 03:00	0	0	12	53
T. Lomnica - Štart	107	141	14.08. 23.08.	183	14.08. 02:00	1	0	20	80
Štrbské Pleso	114	150	23.08.	176	13.08. 17:00	0	0	22	71
Skalnaté Pleso	113	144	23.08.	174	14.08. 01:00, 02:00	0	0	22	87
Solisko	116	144	23.08.	188	13.08. 22:00	3	0	20	-
Lomnický štít	124	153	13.08.	195	14.08. 00:00	8	0	28	198

stations typical ozone diurnal variation shows minimum concentration during nights and peaks at noon, while peak is higher in urban, and on the other hand minimal ozone level in rural sites. Conversely, the highest ozone concentration with minimal daily amplitude appears during night hours at

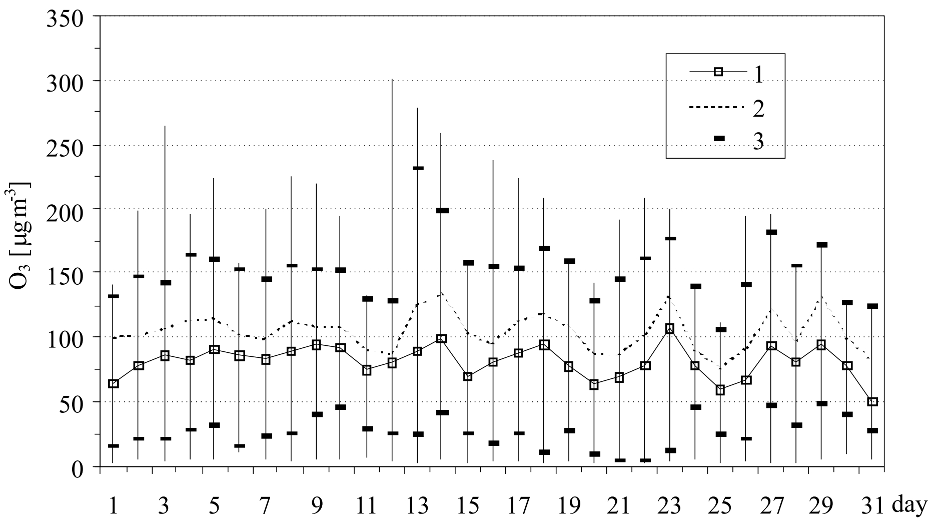

Fig. 3. Ground level ozone concentrations [μg m^{-3}] at urban and rural ozone monitoring stations in Slovakia in August 2003: 1 – daily mean, maximal and minimal hourly concentration O_3 at urban stations, 2 – daily mean at rural stations, 3 – maximal and minimal hourly concentration O_3 at rural stations.

rural stations situated in highland and mountain. In turbulent layer over hill-tops the night-time decrease in ozone is replaced by relatively ozone rich air from above. Daily ozone pattern is generally comparable over a wide geographical area and is associated with: photochemical activity in relation to air temperature and solar radiation daily course, entrance of ozone from free troposphere due to turbulent mixing in the planetary boundary layer and dry deposition. Detailed interpretation is given by *Coyle et al. (2002).*

The ozone monitoring network includes 12 urban and 2 suburban stations in altitude range from 136 to 464 m a.s.l. Background ozone concentrations are observed at 12 rural sites, most of them in highlands and mountain environment - typical cover of Slovakia. In the High Tatras, natural and preserved area, there is a vertical profile elevated from 706 to 2635 m a.s.l. The relationship between mean monthly ozone concentration and altitude in August 2003 is illustrated in Fig. 5. Line 1 shows the slightly decreasing tendency of ozone concentration with altitude for urban stations. At NW upland urban sites e.g. Ružomberok the influence of heatwave flow was probably less intensive than SW lowland urban locality, and there the effect

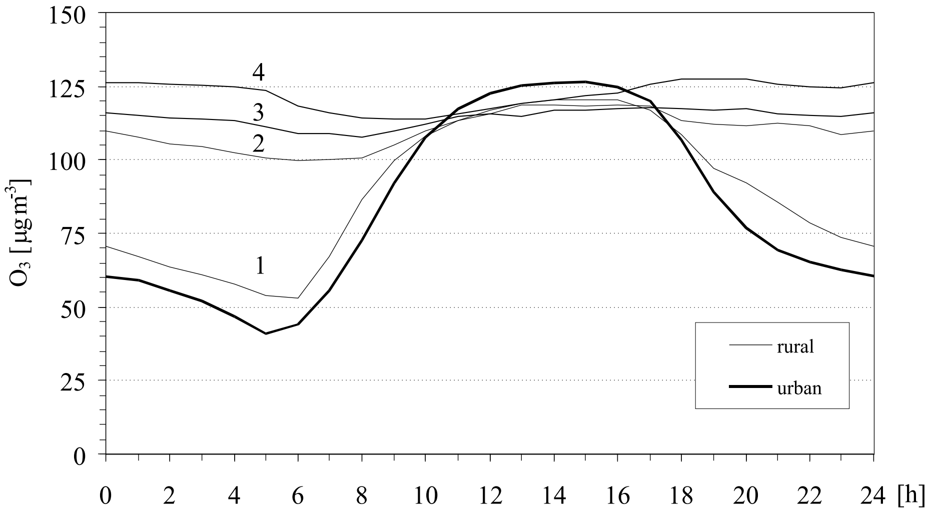

Fig. 4. The daily course of ground level ozone concentrations [μg m^{-3}] at urban and rural stations in Slovakia in August 2003: 1 – altitude 300-1000 m a.s.l., 2 – altitude 1200-1400 m a.s.l., 3 – altitude 1800-1900 m a.s.l., 4 – altitude 2000-2635 m a.s.l.

of emission decrease appears to be proved in August 2003. Variability of mean monthly values for rural sites is higher, although up to altitude 800 m a.s.l. ozone level around 90 μg m^{-3} is relatively stable. Ozone concentration significantly increased with elevation from 800 to 1400 m a.s.l. and indicates ozone accumulation layer over rural sites. The High Tatras vertical profile (line 2) shows rise of mean monthly values from 82 μg m^{-3} in Stará Lesná to 124 μg m^{-3} at Lomnický štít. Fig. 5 indicates special discrepancies. First, comparable values of the highest monthly O_3 mean around 120 μg m^{-3} occurred simultaneously at SW lowland suburban Bratislava-Koliba and N mountain rural Lomnický štít sites. Second, clear different ozone values 127 μg m^{-3} and 255 μg m^{-3} occurred at lowlanded suburban stations Košice-Veľká Ida and Bratislava-Koliba, respectively. *Syri et al. (2001)* consider the urban NO_x emissions as a significant explanatory factor in differences between urban and nearby rural ozone levels. Stationary sources situated in Košice and Bratislava lowland urban localities produce relevant NO_x emissions (Tab. 1). Dominant production of NO_x emission (21%) in Košice locality suggests high potential for ozone photochemistry

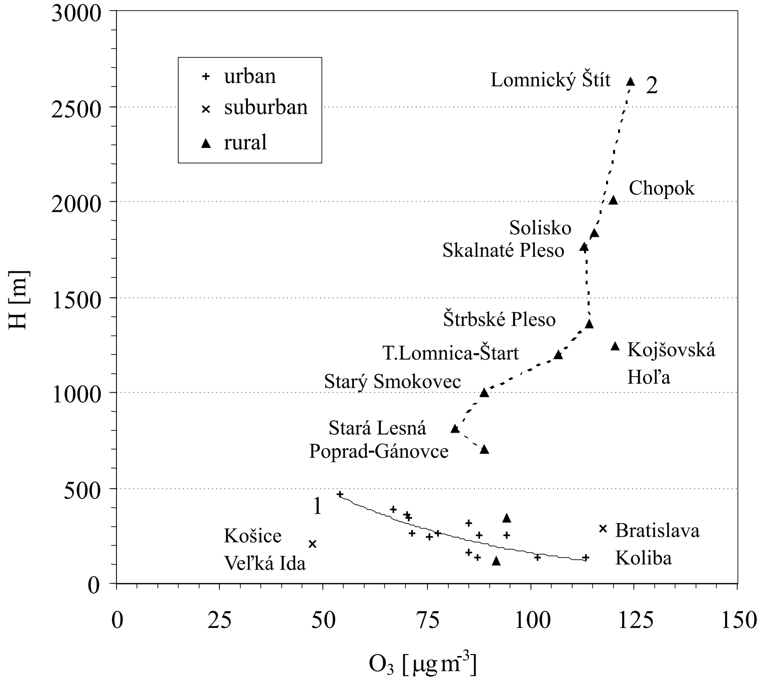

Fig. 5. Vertical profile of ground level ozone concentrations [μg m^{-3}] at urban, suburban and rural stations in Slovakia in August 2003: 1 – urban stations, 2 – the High Tatras stations.

production mainly over suburban site Košice-Veľká Ida. Measured ozone concentrations do not correspond with NO_x production. Considering the general decreasing trend of ozone precursor emissions in Slovakia, high level ozone occurred mainly in SW lowland urban area, while N mountain rural area in August 2003 can be attributed to the arrival of precursors transported from mid and long distances in extremely warm air flow from SW Europe.

Satellite observations enable us to obtain global information on the amounts and distribution of key atmospheric constituents. Global satellite SCIAMACHI (The SCanning Imaging Absorption SpectroMeter for Atmospheric ChartographY) images of tropospheric NO_2 column clearly mark the region with the abundance of NO_2 in the troposphere: SE North America, Central South America, SW Europe, South Africa, China and Japan in August 2003 (http://www.temis.nl/airpollution/no2.html). Situation for the European region indicates the high NO_2 density over urban and vicinity areas of metropolis and industrial cities such as London, Munich, Milano, Prague, Vienna. According to *Li et al. (2002)* higher pollutant density over Western Europe can be associated with transatlantic transport. North American anthropogenic emissions enhance the surface ozone in continental Europe by 10-20 μg m^{-3} during the transatlantic transport events; the transport in the boundary layer and the subsidence from the free troposphere both are important mechanisms. These events tend to occur when ozone concentrations over Europe are about 100-120 μg m^{-3}, i.e., at the threshold of the European ambient air quality standard, and appear to contribute to higher frequency of exceedance. The density of tropospheric NO_2 columns over Slovakia was lower than that over Western Europe. Distribution of NO_2 pollution shows that SW Slovakia, where the capital city Bratislava is situated, and the locality of the High Tatras were significantly influenced. Bratislava, together with conterminous Vienna and Budapest, represents the Danube metropolis triangle area with the potential of highly polluted environment due to urban and industrial emissions. The High Tatras is an area with natural ecosystems, forests, far from urban and industrial areas and away from local emissions. Observation of tropospheric NO_2 columns matches the measured ground level ozone concentration over Slovakia in August 2003 and suggests the relevant transboundary import of ozone and ozone precursors to Slovakia.

4. Conclusion

In this study, statistical characteristics of ground level ozone are analysed according to the field monitoring data in urban, suburban and rural sites in Slovakia during August 2003.

At the most of ozone monitoring stations in Slovakia extraordinary mean hourly O_3 concentrations of 127 to 301 μg m^{-3} were recorded on 12-14 August 2003. Extreme high O_3 concentrations occurred at the SW lowlanded urban stations in Bratislava and at the mountain rural station Lomnický štít. It corresponds to the frequency of excess over the ambient air quality standard. Alert threshold $IH_{1h} = 240$ μg m^{-3} 6 times only in Bratislava and information threshold $IH_{1h} = 180\ \mu g\ m^{-3}$ also at rural mountain stations at altitude 1100-2635 m a.s.l. were frequently exceeded. The target value 8h mean $IH_{8h} = 120\ \mu g\ m^{-3}$ was exceeded also in Bratislava and Lomnický štít over more than 25 days.

Mean daily ozone concentrations at rural stations were higher than at urban stations, while mean hourly ozone concentration amplitude at urban stations was substantially higher than at rural stations. Different variability reflects the daily ozone pattern. The daily course of ozone concentration depends on the type of station, altitude, daily course of air temperature, and solar radiation, entrance of ozone from free troposphere and its loss due to dry deposition.

Favourable weather conditions in abundance of NO_x and VOC support the photochemistry ozone process yielding to the O_3 concentrations episode. Extreme high temperatures (positive deviations of 2.3° C to 4.8° C above the long-term value), precipitation (deficit 45 mm), low air relative humidity (11% to 18% under normal) and sunshine duration (141% to 205%) were recorded in Slovakia in August 2003. The significant temperature elevation occurred mainly in SW Slovakia. Maximum temperature of 38.8° C was on 13 August 2003 in Bratislava. During 12-13 August also extreme high ozone concentrations in Bratislava were recorded. Summer ozone maximum in August 2003 appears to be associated with special ozone production and distribution due to the unusual warm weather situation and the transport of abundant ozone precursors over Europe. The decreasing trend of NO_x pollutant suggests the ozone and precursors transboundary transport into Slovakia.

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