Analysis of ambient ozone in a foothill area in the Western Carpathians

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Abstract

Kellerová, D., Janík, R. 2014. Analysis of ambient ozone in a foothill area in the Western Carpathians. *Folia oecol.*, 41: 146–152.

This work analyses the ambient ozone concentrations measured during 1999–2008, parallel on two research plots differing in their vegetation cover (beech forest stand and open plot), situated in a rural area. There was detected a difference in the ozone concentration values between the two plots due to their spatial arrangement, but this difference was not significant. On the other hand, a noticeable statistically significant difference in ozone concentrations and differences in selected climatic variables were observed. The influence of average temperature and of rainfall sum was very significant, at $p < 0.0001$. The variability of the measured ambient ozone values ranged from 50.01% in the stand to 57.36% on the open plot. Ozone maxima occurred frequently, mainly after the year 2004. The increase in ozone concentrations, primarily in form of extreme events, means a serious risk for the environment.

Keywords

ambient ozone, forest beech, open plot, rural area, statistical analysis, the Western Carpathians

Introduction

Several factors, both anthropogenic and natural, influence forest ecosystems in central Europe and cause important changes to these ecosystems, their climate included. The former status quo in the emitted and airborne pollution has been changed considerably. Comparing to the past, the effects of acid substances have been mitigated (Fleischer et al., 2009). On the other hand, the trees, both broadleaved and coniferous, became affected negatively or even destructively by ambient ozone present in high concentrations (Muzika et al., 2004; Karlsson et al., 2007; Sanz et al. 2007; EEA, 2010). Generation, formation and accumulation of ambient ozone is a process comprising several levels, and it is determined by various conditions and interactions. The dominant mechanism of pollution transport to Slovakia is transboundary. The ozone precursors are transported in the free troposphere, in horizontal direction. They then descend vertically to the lower layers and mix with nitrogen oxides (NO_x) and volatile organic compounds (VOCs) coming from local anthropogenic and natural sources. The main local source of nitrogen oxides is car transport, expanding dynamically. Another important source is NO_x emitted from soils, in the Slovak Republic estimated up to 15% of the total amount emitted in Europe (SHMI, 2008). Not only in Slovakia, but also in Europe, the trend of concentration of nitrogen oxides does not decrease comparatively to the sulfur oxides (Smidt et al., 2010; SHMI, 2010).

Forests, the dominant source of biogenic volatile organic compounds, represent 41% of the territory of Slovakia. Accordingly, they can produce VOCs (primarily terpenes and isoprenes) in amounts sufficient, under certain circumstances, to promote the local development of ground level ozone.

In the central Europe, the role of nitrogen oxides interacting with anthropogenic and biogenic organic substances and with solar radiation is much more important than formerly supposed. Further significant factors are: the daily wind course, thermal circulation in rugged terrain, deposition onto the surface and interaction among all ozone layers (SHMI, 2006; Šec et al., 2007; Castell et al., 2008).

In this context, there is an urgent need to investigate the relation between the prominent pollutant – ambient ozone, and the ecosystem and its surroundings influenced by this pollutant (Paoletti et al., 2010; Serengil, et al., 2011; Matyssek et al., 2012).

The aim of the experiment was to assess the actual condition and the differences in the ozone distribution patterns for two different research areas in rural environment. We analysed the extent to which the ozone concentrations were dependent on the local temperature and precipitation.

Material and methods

The results of the experiment demonstrate the differences in the ground level ozone concentrations measured simultaneously on two different plots situated at the same altitude. The first plot was covered with a connected beech forest stand; the second was an open area, without forest cover. We analysed the differences related to the actual local situation concerning the airborne pollution, microclimate and meteorology. Our question was whether a forest stand could affect ground level ozone concentrations as it is known for other common pollutants for which forests act as effective filters capturing undesirable harmful substances. We investigated spatial and temporal distribution, differences in seasonal and annual concentrations, and exceeding-limits events. This research's duration was 10 years, so the data obtained could be used to found out if the concentration development showed some trends and dynamics. The parameters were explored and characteristics were processed statistically.

Study site

The experimental plots are situated in the Kremnické vrchy Mts (φ = 48°38´10´´ N, λ = 19° 04´08´´ E), Slovakia, Central Europe. The plot with dominant beech (94.7%) is located at 480–510 m a.s.l. (Barna and Dobrovič, 2010). The slope is west oriented, with an inclination of 30 to 36% (Schieber, 2007). The stand age at the beginning of the experiment was 100–110 years. The open plot is on the foothills of a beech-covered slope, at ca 480 m a.s.l. The distance between the two plots is ca 100–150 m.

The climate in the Kremnické vrchy Mts is moderate warm and moderate humid. The long-term mean annual air temperature is 8.2 °C, in the growing season 14.9 °C; the annual rainfall total ranges from 510 mm to 1,040 mm, in the growing season from 160 to 530 mm (Kellerová and Dubová, 2002; Janík, 2006).

As for the airborne pollution, the research plots investigated are outside the direct impact of polluting substances; neither are they severely affected by the transboundary pollution transport. Three stationary energy production units in the Zvolenská basin close (ca 10 km) to the research site can influence the plots in case of "assistant" meteorological conditions. We were doing our research in the spring and summer when the meteorological, conditions in the basin were mostly favourable for ozone generation (SHMI, 2012). It is supposed that the ozone generation in the locality Kremnické vrchy may be elevated due to precursors (isoprenes and terpenes) coming from the natural sources – forests. Input of further airborne pollutants into the Kremnické vrchy Mts is discussed in Dubová and Bublinec (2006) and Janík et al. (2011).

The temperature data were taken from the SHMI station Sliač (φ = 48°38´33´ N, λ = 19°08´31´´ E), located at 313 m a.s.l. The climatic characteristics are reported for the period 1999 to 2008.

Monitoring of O_3 concentration

The possibilities for monitoring of airborne pollutans in submountain conditions are limited, so the data are supplemented with figures obtained by statistical processing or with information assembled by using passive samplers. The passive samplers are easy to operate. They only demand low costs, work without an energy source, and enable precise discrimination of risk territories with regards to the potential damage to the ecosystems. The methods are progressively improving, simplified, and their results can be compared with the results obtained by using continual analysers (Gerosa et al., 2001; Cox, 2003; De Vries et al., 2003; Bytnerowicz et al., 2004; Šrámek and Novotný; 2009). The drawback of these methods is that they are not suitable for 24-hour monitoring of the dynamics of the ozone concentrations.

The Werner's method (1991) for ozone quantification is based on the selective reaction between a layer of indigo applied on a filter paper and the atmospheric ozone. Such papers are exposed in the field for 7–10 days, during growing season (April–September). The passive ozone samplers are placed on each plot, in pairs, at 1.5 m above the ground. After the exposition, the papers are extracted with ethanol in the laboratory. The reaction between indigo and ozone results in the production of isatin, which is indicated by yellowing of the test papers. The content of isatin is determined by spectrophotometry at a wave-length of 408 nm. The final value of the extinction is proportional to the isatin content and, consequently, also to the ozone content. The measured extinction values reflect the ozone sums according to the calibration curve. The ozone concentrations are given in standard units of ppb per a day or in μg m^{-3} per a day. From the daily values, we calculated the monthly and annual characteristics. The indigo papers were exposed in an equipment consisting of a roofed stand and a perforated cylinder (passive sampler) into which the papers prepared in the laboratory were fixed. The detail description is in Kellerová (2002).

The precipitation was captured with funnels (each 660 cm^2 in area) into closed collectors. The collectors were spaced regularly across the stand (by 10 ts) and across the adjacent open area. The samples were taken at regular intervals and after each precipitation event. They were then subjected to chemical analysis and processed in the laboratory. The representative samples were defined as mixture from all the collectors in the stand and in the open plot.

Statistical characteristics of measure and position were calculated and the tests were performed with using the software package Statistica v 7. The normalness of distribution was tested with the Shapiro-Wilk W test. The significance of differences of basic sets between the monitoring plots was evaluated with the Student's t-test for independent variables. The results were also verified with using the software package Statistica.

Results and discussion

Our research purpose was to examine the differences in the ambient ozone concentrations between two researches plots situated in the Kremnické vrchy Mts, Slovakia, Central Europe: one representing a beech forest stand (*Fagus sylvatica L.*) and the other an open area, outside the forest. We analysed the extent to which the ozone concentrations were dependent on the local temperature and rainfall. The measurements were carried out during the growing season, from April to September, when the ozone concentrations are generally higher than in colder months.

The first comparison of the temporal and spatial patterns of ozone concentration between the plots resulted in finding that in the first half of the research period 1999–2003, the ozone concentrations in the beech forested plot were higher (38 $\mu g\ m^{-3}$) than on the open plot (32 $\mu g\ m^{-3}$). In the second half of the experiment, 2004–2008, this trend was only observed in 2005 and 2007 (Fig. 1). Nevertheless, in any case, the differences in ozone concentrations between the two plots were not statistically significant.

To sum up, after 2004, the ozone concentrations increased from 39 to 55 $\mu g\ m^{-3}$. The difference in ozone concentrations between 1999–2003 and 2004–2008 was tested statistically, and it was found very significant (Table 1). The cause may be due to the high air temperature and low rainfall, mainly in years 2006–2008. The air temperature mean we obtained in the growing season was 16.0 °C which is evidently higher than the corresponding long-term mean of 14.5 °C reported from Sliač for the period 1951–1980.

The clearly highest average values of the ozone concentrations on the two experimental plots in the Kremnické vrchy Mts were recorded in 2007 (beech stand 88, open plot 75 $\mu g\ m^{-3}$). In this year, there was also measured the highest average air temperature in the growing season (16.2 °C). According to the evaluation by SHMI (2008), the photochemical activity in year 2007 was above average, and the ozone concentrations in rural areas were by 30–40 $\mu g\ m^{-3}$ higher than the values corresponding to the same altitudes in years with lower photochemical activity.

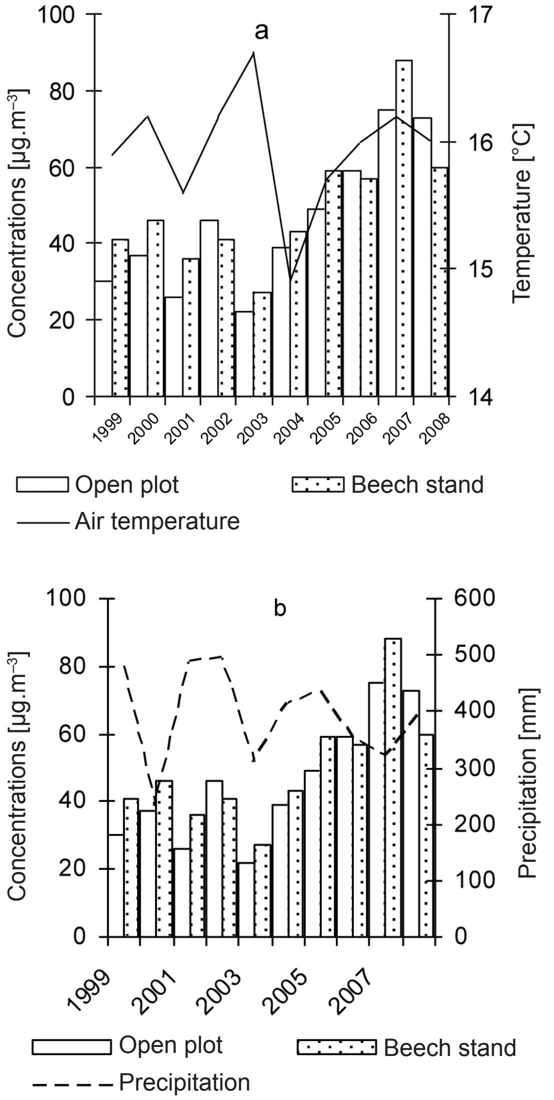

Fig. 1. Average annual ambient ozone concentrations on two-sample plots (open area and beech forest), compared to air temperature (a) and rainfall (b).

In summary, the variability of our ambient ozone concentrations ranged from 50.01% in the stand to 57.36% on the open plot (Table 2). Our research results do not allow us to judge about time trends, as our inter-annual variability in ozone concentrations was high,

Table 1. T-test and p values for open plot, beech stand, air temperature and precipitation total in the Kremnické vrchy Mts in years 1999–2008

	Open plot	Beech stand	Air temperature	Precipitation total	Open plot	Beech stand	Air temperature	Precipitation depth
	t-values				p<			
Open plot	–	–0.526	5.045***	–12.255***	–	–	–	–
Beech stand	0.526	–	6.267***	–12.147***	–	–	–	–
Air temperature	–5.045***	–6.267***	–	–13.669***	–	–	–	–
Precipitation depth	12.255***	12.147***	13.609***	–	–	–	–	–
Open plot	–	–	–	–	–	0.605	0.00001	0.00000
Beech stand	–	–	–	–	0.605	–	0.00001	0.00000
Air temperature	–	–	–	–	–0.0008	–0.00001	–	0.00000
Precipitation total	–	–	–	–	0.00000	–0.00000	0.00000	–

Marked differences are significant at $p < 0.05$.

Table 2. Descriptive statistics of ambient ozone in the Kremnické vrchy Mts in years 1999–2008

Statistical parameters	Open plot	Beech stand
Valid	48.00	54.00
Mean	48.11	51.39
Standard error	3.98	3.49
Standard deviation	27.59	25.72
Minimum	8.00	10.00
Maximum	144.00	140.00
Range	136.00	130.00
Variance	761.46	661.29
Lower Quartile	25.50	32.00
Upper Quartile	64.00	64.00
Coefficient of Variation	57.35	50.05
Sum	2,309.00	2,775.00

and the trend evaluation in this area requires much longer time series than the series we have recorded till now (Jonson, et al., 2006).

Figure 2 illustrates that until 2005, the measured maxima had exceeded the limit of 65 µg m^{-3} only in three cases, whereas later all the maxima were above this limit value.

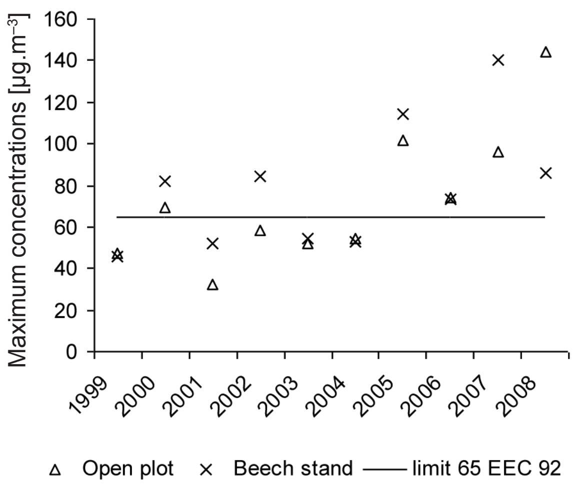

Fig. 2. Maximum ozone concentrations on two plots in the Kremnické vrchy Mts, representing open area and beech forest in summer period (April–September) on 1999–2008. Maximum concentrations compared with ecological limit for vegetation (65 µg m^{-3}).

The ground level ozone concentrations and short-term limit values of airborne pollutants were compared with the value of 65 µg m^{-3} (32.5 ppb day^{-1}) approved as ecological limit for the 24 h average concentration of this substance by the EU in 1992 (EEC 1992). The events of exceeding this limit in the stand were by 10% more frequent than on the open plot. In total, the highest ground level ozone concentrations were measured in 2007 (140 µg m^{-3}) in the stand and in 2008 (144 µg m^{-3}) on the open plot, the lowest in 2005 (8 µg m^{-3}) on the open plot and (10 µg m^{-3}) in the stand.

From the viewpoint of seasonal dynamics, in the late summer (July, August), higher ambient ozone concentrations were recorded on the beech-forested plot; in the spring and early summer (April, June), on the open plot. The year 2004 seemed to change the former trend, probably owing to the meteorological situation during the measurement. This can be concluded from the influence of selected climatic variables (average temperature and rainfall sum) on the ozone concentrations. The changes in meteorological parameters, especially the increase in air temperature linked with the global climate change, become evident also according to recent research (Bičárová and Fleischer, 2007). Another cause may be the tree harvest in close proximity of our beech stand, which could influence the microclimate and site conditions in this stand. The rise in ozone concentrations and in air temperature recorded recently in this submountain area may be warning, despite its values are not as extreme as those reported from other regions of Slovakia and from Central Europe (Hůnová et al., 2012; Šrámek et al., 2007; Melkonyan and Wagner; 2013). The conditions promoting ozone formation depend on the local situation in airborne pollutants and on the local microclimate and meteorology during anti-cyclones expanding over big areas. High ground level ozone concentrations developed mainly during episodes with abundant photo-chemical smog linked typically with stagnant air, sunny summer heat and low air humidity.

The number of episodes in exceeding the limit was somewhat higher in the stand than on the open plot. This suggests the presence of further agents acting synergically and influencing ozone concentration in forest stands. Biogenic emissions of volatile organic compounds and nitrogen oxides from natural sources can play an important role in the ozone formation over Europe. This role may be much more important than supposed so far. Such ozone precursors as terpenes and isoprenes are forest products. The major part of isoprenes and more than one half of terpenes are emitted from May to August, and the emission of these substances rises exponentially with raising temperature (SHMI, 2008). The beech forests contribute to the total biogenic VOC emissions by ca 10% (Bičárová, 2008). The primary local sources are supposed: anthropogenic emissions from car traffic, recently built industrial park and local combustion of solid fuels. The diverse terrain topography enables, under specific conditions, mixing of ozone and its precursors with the valley circulation, which also increases the local ozone concentrations (SHMI, 2006).

Given that research lasted 10 years, we may outline certain trends in ozone concentrations. Figure 3 shows that the trends on both plots were rising significantly.

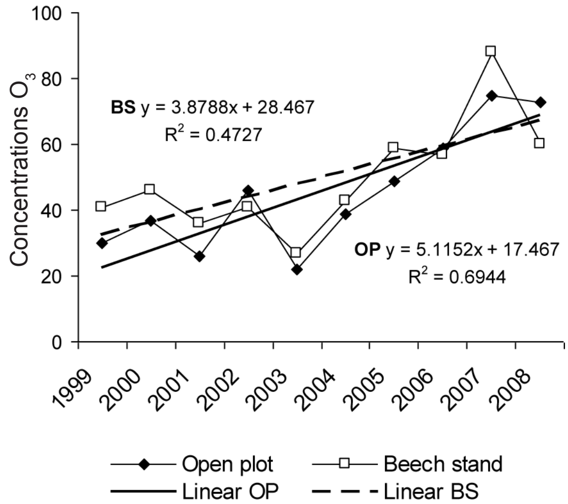

Fig. 3. Linear trend and temporal distribution of ozone concentrations on research plots in the Kremnické vrchy Mts.

Conclusion

We analysed and evaluated ambient ozone concentrations measured over the period 1999–2008 on two research rural plots situated at the same altitude but differing in their vegetation cover.

The statistical analysis revealed that the differences in the ozone concentration values between the two plots were not significant. In the case of ambient ozone, the forest did not act as a filter for trapping the pollutants. Contrarily, the forest contributed to the pollution, due to the precursors produced by the forest itself. On the other hand, the pollution manifested distinct temporal variability, because the average monthly ozone concentration values were more often high, even above limits, and the inter-annual differences were impressive just after the year 2004. It is not easy to draw conclusions for the temporal trends, because the inter-annual variability of the ozone concentration was relatively high and because the judgment about the trends needs temporal series much longer than the series we dispose with.

Statistically very significant was found the influence of average temperature, rainfall sum and clearly also tree felling in the surrounding stands. In terms of seasonal dynamics, higher ambient ozone concentration values in the forest stand were recorded in July and August, on the open plot in April and June. The rise in ozone concentration, primarily in the form of extreme events, seems to be a serious risk for the environment.

Acknowledgement

This work was supported by the Slovak Research and Development Agency under the contract No. APVV-0429-12 and by the Scientific Grant Agency of the Ministry of Education of the Slovak Republic and the Slovak Academy of Sciences (Projects No. 2/0089/14, 2/0027/13, 2/0053/14 and 2/0113/12). We also acknowledge D. Kúdelová for preparing the English text.

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Received March 3, 2014
Accepted September 8, 2014