# Ambient ozone phytotoxic potential over the Czech forests as assessed by AOT40

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Ambient ozone  $(O_3)$  represents one of the most prominent air pollution problems in Europe. We present an analysis on  $O_3$  with respect to its phytotoxic potential over Czech forests between 1994 and 2008. The phytotoxic potential is estimated based on the exposure index AOT40 for forests calculated from real-time monitoring data at 24 rural sites. Our results indicate high phytotoxic potential for most of the Czech Republic (CR) with considerable inter-annual and spatial variability. The highest AOT40 values were 38-39 ppm  $\cdot$ h. The critical level for forest protection (5 ppm  $\cdot$ h) was usually exceeded early in the growing season, generally in May. In years with meteorological conditions conducive to ozone formation, the critical level was exceeded by 5-7 folds as compared to years with non-conducive conditions; nevertheless, all sites consistently exceeded the critical level since 1994. In the extremely hot and dry year 2003, the critical level for forests was exceeded over 31 % of the Czech forested area. More research is needed to translate these exceedances into forest injury in the CR.

Keywords: Ozone, AOT40, Time trends, Spatial patterns

# Introduction

Ambient ozone (O<sub>3</sub>) has been a widely studied air pollutant for many years due to its potential toxicity for all living organisms (Felzer et al. 2007, Cape 2008, Iriti & Faoro 2008). It is an important gas playing a key role in atmospheric chemistry (Seinfeldt & Pandis 1998). It contributes to the oxidative power of atmosphere which is essential for scavenging many pollutants from the air. Moreover, due to its absorption-radiation abilities, O<sub>3</sub> is an important greenhouse gas (Singh & Fabian 2003, IPCC 2007). There are important mutual interactions between O<sub>3</sub> and climate change (Isaksen 2003), which are not fully understood yet. The urgent need to address the knowledge gaps in interactions between air pollution, climate change and forests has been recently stressed (Serengil et al. 2011, Matyssek et al. 2012).

Ozone represents one of the most promi-

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nent air pollution problems in Europe (De Leeuw & De Paus 2001, EEA 2010). Environmental O<sub>3</sub> quality standards are exceeded over vast areas of Europe (Horálek et al. 2007, 2009). Due to its phytotoxicity, O<sub>3</sub> is still considered to be the most important air pollutant for forests (Paoletti et al. 2010). Due to the fact that O<sub>3</sub> is a secondary pollutant formed from precursors during complex photochemical reactions and to the highly non-linear nature of O<sub>3</sub> chemistry, it is and will be very difficult to decrease its ambient concentrations (Seinfeldt & Pandis 1998).

Comparison of O<sub>3</sub> levels with those measured a century ago indicates that current levels have increased by approximately two times. European measurements between 1850 and 1900 were found to be in the range of 17-23 ppb (Bojkov 1986). Modern day annual average background O3 concentrations over the mid-latitudes of the northern Hemisphere range between approximately 20-45 ppb, with variability influenced by geographic location, altitude and extent of anthropogenic impacts (Vingarzan 2004). Generally, three types of patterns in ambient  $O_3$  have been apparent recently: (1) an increase in the extent of O<sub>3</sub> impact and the forest areas at risk; (2) a decrease in maximum 1-h O<sub>3</sub> concentrations, at least in the northern hemisphere countries which have introduced O<sub>3</sub> precursor control programs; and (3) an increase in background  $O_3$  concentrations over much of the world (Percy et al. 2003). It is not an easy task to assess the time trends of O<sub>3</sub>: the inter-annual variability is fairly high, so that long time series, which mostly are not available (Jonson et al. 2006), are needed to detect trends.

In the Czech Republic (CR), ambient air pollution has been perceived as a major environmental problem since the 1950s, particularly due to extremely high emissions of SO<sub>2</sub> and particulate matter from large powergenerating sources (Moldan & Schnoor 1992). Pollution in the form of surface  $O_3$ was recognized as an issue as late as in the 1990s. Its levels are regularly measured within the framework of a national ambient air quality network run by the Czech Hydrometeorological Institute (CHMI) since 1993. Ozone levels are relatively high, and the limit values (EC 2008) over vast regions are frequently exceeded (Hůnová 2003). Mean O<sub>3</sub> concentrations during the growing season at rural sites range between 30 and 45 ppb in years with low O<sub>3</sub> levels (as in 2001 and 2008) and between 35 and 60 ppb in years abundant in O<sub>3</sub> (as in 2003). Peak 1-h mean O<sub>3</sub> concentrations reached 110 ppb in 2003, 90 ppb in other years (Hůnová et al. 2010).

Based on long term real-time monitoring, we present an analysis of O<sub>3</sub> time trends and spatial variability with respect to its phytotoxic potential over Czech forests between 1994 and 2008. Out of the two approaches (concentration-based and flux-based) developed for O<sub>3</sub> risk assessment by UN/ECE (2004), we used the concentration-based approach and applied the exposure index AOT40. We are fully aware of increasing number of studies promoting the flux approach as more scientifically sound (e.g., Ashmore et al. 2004, Matyssek et al. 2007, Tuovinen et al. 2009) and of numerous criticisms of AOT40 concept and robustness (e.g., Sofiev & Tuovinen 2001). Moreover, O<sub>3</sub> stomatal flux based indexes are reported to outperform AOT40 for explaining the biological effects such as biomass reduction and leaf visible injury (Karlsson et al. 2007). From a practical point of view, however, it is obvious that the exposure index has the advantage of relative simplicity, and in regions which are not under stress by drought which is generally the case of the Czech mountain forests - the areas at risk indicated by exposure index and stomatal flux are not likely to differ substantially. However, even in Japan, where annual precipitation is usually very elevated, the flux approach was recommended when VPD is a limiting factor to stomatal uptake (Hoshika et al. 2011). The AOT40 has an advantage of relatively simple calculation based on ambient O<sub>3</sub> concentration data, while modeling of stomatal flux is much more complicated (Tuovinen et al. 2009). Modeling of stomatal flux needs  $O_3$ concentrations and data for stomatal conductance to be measured or modeled. In case the measured data are not available (which is the case for the CR), we are likely to intro-

	Tab. 1	l - 1	Sites	used	for	the	analysis	ranked	l according	to	decreasing	altitude
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Commentance	Site Name	Site Identifier	Altitude			
Country	Site Name	(ID)	(m a. s. l.)			
Czech	Churanov	1	1118			
Republic	Serlich	2	1011			
	Krkonose-Rychory	3	1001			
	Prebuz	4	904			
	Bily Kriz	5	890			
	Rudolice v Horach	6	840			
	Hojna Voda	7	818			
	Sous	8	771			
	Cervena	9	749			
	Primda	10	740			
	Svratouch	11	735			
	Jesenik	12	625			
	Stitna n. Vlari	13	600			
	Sneznik	14	590			
	Kostelni Myslova	15	569			
	Kosetice	16	535			
	Kocelovice	17	519			
	Ondrejov	18	514			
	Valdek	19	438			
	Kucharovice	20	334			
	Tusimice	21	322			
	Lom	22	265			
	Mikulov-Sedlec	23	245			
	Studenka	24	231			
Poland	Sniezne Kotly	25	1490			
	Czarna Gora	26	1133			
	Czierniawa	27	645			
	Jeleniow	28	244			
Germany	Fichtelberg	29	1213			
-	Carlsfeld	30	896			
	Zinnwald	31	877			
	Schwartenberg	32	787			

duce large uncertainties into the calculation. The shortcomings encountered in modeling  $O_3$  flux are discussed by Tuovinen et al. (2009).

# Methods

## Ozone data

For our analysis, we used the data measured within the framework of the nation-wide ambient air quality monitoring network operated by the Czech Hydrometeorological Institute (CHMI). Ambient O3 monitoring over the CR has evolved significantly since its beginning in 1993. The number of sites within this network has largely increased from an original 16 in 1993 to 55 presently covering rural, mountain and urban areas (Ostatnická 2011). The ambient O<sub>3</sub> concentrations were measured by real-time analyzers (Thermo Environmental Instruments TEI, M49) using UV-absorbance, a reference method in the EC (2008). Standard procedures for quality control and quality assurance (EC 2008) were applied. We only considered sites with relatively large spatial representativeness, *i.e.*, those classified as rural according to the EoI classification (EC 1997). Overall, we used 24 Czech sites. For mapping purposes, four additional German and four Polish sites were included (Tab. 1, Fig. 1).

# Exposure index AOT40Forest

We analyzed the annual trends for selected

sites representing the principal Czech mountain regions (1. Churanov - the Sumava Mts., 3. Krkonose-Rychory - the Krkonose Mts., 5. Bily Kriz - the Beskydy Mts., 6. Rudolice v Horach - the Krusne hory Mts., 8. Sous the Jizerske hory Mts., 12. Jesenik - the Jeseniky Mts.), a regional site considered to represent the CR background (16. Kosetice the Czech-Moravian Highlands), and a regional site representing the relatively warm lowlands in Southern Moravia (23. Mikulov-Sedlec). For the assessment of ambient ozone phytotoxic potential for forests, we applied the AOT40 approach (Fuhrer et al. 1997, UN/ECE 2004). The exposure index AOT40 was calculated according to eqn. 1 (see below). For practical reasons, we considered the daylight hours between 8 a.m. and 8 p.m. Central European Time (EC 2008).

$$AOT40 = \sum_{i \in V} \sum_{j=1}^{n} \sum_{k \in D} \left( c_{ijk} - p \right)$$

where  $c_{ijk}$  is the ground-level O<sub>3</sub> concentration measured in the *i*-th month, *j*-th day and *k*-th hour; *p* is the threshold concentration (40 ppb); *V* is a set of the months of the growing season (April-September); *D* is a set of daylight hours, defined as those hours with a mean global radiation of 50 W m<sup>-2</sup> or greater; and *n* is the number of days in the month.

We carefully checked the data coverage for calculating AOT40. When all possible data were not available for calculation of the AOT40 due to monitoring gaps (Tab. 2), we used the correction factor recommended by EC (2002 - eqn. 2):

$$AOT40_{(estimated)} = AOT40_{(calculated)} \cdot a/b$$

where a is the total possible number of hours, and b is the number of measured



Fig. 1 - Sites with on-line monitoring of ambient ozone used for AOT40 mapping.

**Tab. 2** - Data coverage for calculating AOT40Forest expressed as percentage of 1-h mean  $O_3$  concentrations available over the growing season (1.4-30.9). The site ID corresponds to the site name in Tab. 1.

Country	Station ID	Data coverage (%)														
Country	Station ID	1994	1995	1996	1997	1998	1999	2000	2001	2002	2003	2004	2005	2006	2007	2008
Czech	1	-	70	98	99	99	99	99	99	98	96	90	100	100	99	100
Republic	2	-	33	94	93	94	92	93	98	96	98	98	98	99	100	100
-	3	-	-	96	87	98	98	92	92	98	99	98	82	89	98	100
	4	61	91	99	98	98	98	99	100	97	99	99	98	96	99	99
	5	97	97	88	96	93	98	99	99	99	98	99	97	100	99	99
	6	-	-	98	98	98	90	99	100	98	98	97	97	93	100	100
	7	-	99	82	91	88	99	99	92	96	90	98	100	96	99	100
	8	88	99	94	97	99	98	99	100	99	97	95	96	99	97	97
	9	-	-	-	-	-	-	-	-	-	-	99	99	96	88	98
	10	-	81	99	97	98	95	93	98	94	97	96	100	100	100	100
	11	90	88	99	85	98	99	98	96	96	81	99	100	99	100	100
	12	-	99	94	98	99	99	99	99	97	100	99	100	100	96	100
	13	-	-	-	-	-	-	-	-	-	91	98	99	100	100	99
	14	-	95	98	99	96	98	98	98	99	99	32	98	98	100	100
	15	-	-	-	94	98	100	96	97	98	99	78	99	99	99	99
	16	92	94	96	99	99	93	100	97	98	100	99	99	96	100	100
	17	-	-	-	-	-	-	-	-	-	-	98	98	97	100	100
	18	75	98	90	86	94	100	95	97	97	96	91	95	99	100	100
	19	-	-	-	-	-	-	-	-	-	98	81	99	99	99	97
	20	-	-	-	-	-	-	-	-	-	-	-	95	99	100	99
	21	70	94	96	91	100	97	99	98	99	99	99	99	99	100	100
	22	-	-	-	-	-	-	-	-	-	-	-	99	100	100	99
	23	-	-	100	100	96	98	99	100	98	99	98	92	97	99	100
	24	-	-	-	-	95	98	99	99	99	100	100	96	100	100	100
Poland	25	-	-	-	80	95	61	76	48	77	63	65	90	93	94	45
	26	-	-	-	-	98	99	93	48	96	92	92	94	99	87	96
	27	-	-	-	93	89	75	87	48	98	88	92	69	98	99	95
	28	95	93	96	-	-	-	-	-	94	87	84	96	93	100	100
Germany	29	-	-	-	88	90	97	96	98	93	92	91	16	96	98	96
	30	-	-	75	95	97	95	97	97	93	92	-	16	99	100	100
	31	-	-	85	94	96	96	91	95	93	95	-	16	99	97	99
	32	-	-	-	-	95	94	92	94	90	95	-	16	99	99	99

## hourly values.

#### AOT40Forest spatial pattern

The spatial distribution of AOT40Forest was carried out for three years: 2003, 2006 and 2007. We selected 2003 due to its exceptional meteorological conditions (extremely high temperatures spanning long durations), 2006 as an example of a year with high  $O_3$  exposures, and 2007, in contrast, as an example of a year with low  $O_3$  exposures during the growing season.

For mapping, we used a linear regression model with subsequent IDW (Inverse Distance Weighting) interpolation of the residuals. AOT40 calculated for the monitoring sites was used as a dependent variable for the regression model. Orography (altitude) was used as an independent variable. For the spatial interpolation of the residuals, we used the deterministic method IDW included in ArcGIS Geostatistical Analyst (Johnston et al. 2001). The IDW spatial interpolation technique (*e.g.*, Isaaks & Srivastava 1989) estimates the cell values using a weighted linear combination of values measured at several neighborhood sites, where the weight is an inverse function of the distance, according to the following equation (eqn. 3):

$$Z(s_{0}) = \frac{\sum_{i=i}^{n} \frac{Z(s_{i})}{h_{0i}^{\beta}}}{\sum_{i=i}^{n} \frac{1}{h_{0i}^{\beta}}}$$

where  $Z(s_0)$  is the interpolated grid value;  $Z(s_i)$  is the neighboring data point;  $h_{0i}$  is the distance between the grid node and the data point;  $\beta$  is the weighting power (the Power parameter); and *n* is the number of measuring points.

Maps were prepared at resolution 1x1 km. The digital map of Czech forests produced from the European digital land use map (Corine Land Cover 2000 - http://etclusi.eionet.europa.eu/ CLC2000) was used. A spatial categorization of AOT40 was carried out only for forested areas, which form about 33 % of the Czech territory. For interpolation in border areas, we used data from four German sites for the region of the Krusne hory Mts. and four Polish sites for the northern part of the CR. The spatial distribution of the sites was surprisingly even in altitude (Tab. 3); in contrast, it was uneven regarding the geographical distribution (Fig. 1). The sites were concentrated mostly in mountain areas; the densest network was in the Krusne hory Mts., *i.e.*, in the north-west.

#### Ozone precursor data

The data on emission of NO<sub>x</sub>, VOC, CO

**Tab. 3** - Spatial distribution of the monitoring sites (ranked according to the altitude) used for  $O_3$  exposure mapping.

Altitude (m a.s.l.)	Number of Czech sites	Total number of sites (including the sites abroad used for border analysis)					
231-400	5	6					
401-600	7	7					
601-800	5	7					
801-1000	4	6					
1001-1200	3	4					
1201-1490	0	2					
Total	24	32					









Fig. 3 - Average temperature for the CR (source: CHMI).



Fig. 4 - Histogram of 1-h mean  $O_3$  concentrations at selected rural sites (indicated as 1, 3, 5, 6, 8, 12, 16, 23) in the CR, 1994-2008.

were taken from the Register of Emissions and Air Pollution Sources (REZZO), the Czech emission inventory database run by CHMI. REZZO includes information on anthropogenic sources of air pollution, both stationary (categorized as extra large, large, medium and local) and mobile (Ostatnická 2011). The data on emissions of CH<sub>4</sub> were taken from the National Greenhouse Gas Inventory Report (NIR) of the CR (Fott & Vacha 2011) and these include both anthropogenic and natural sources.

# Temperature data

We used the data measured at climatological stations run by the CHMI. Manual climatic measurements were taken by a station thermometer in a standard thermometer screen 2 m above-ground at climatological observation times, i.e., 7 a.m., 2 p.m. and 9 p.m. local mean solar time (LMST). Seasonal mean temperature was calculated based on daily mean temperatures. Daily mean temperature was calculated as an average of the temperatures at the observation times, where the evening observation was used twice. Seasonal mean temperature was calculated from all available stations which had records for the relevant year, *i.e.*, we used about 200 stations per year (Tolasz et al. 2007).

# Results

Fig. 2 and Fig. 3 summarize the trends of factors which substantially influence ambient O<sub>3</sub> levels. Emission of O<sub>3</sub> precursors from Czech anthropogenic sources between 1990 and 2008 decreased by 52 % for NO<sub>x</sub>, by 63 % for non-methane VOCs, by 65 % for CO, and by 42 % for CH<sub>4</sub>. For the period under consideration, i.e., 1994-2008, the emission of NO<sub>x</sub> decreased by 30 %, nonmethane VOCs by 47 %, CO by 58 %, and CH<sub>4</sub> by 23 %. Though somewhat hidden in a seasonal mean, the air temperature variability was fairly high. In 2003, for example, the temperature in the CR was 4 °C and 3.6 °C above the average in July and August, respectively.

The distribution of 1-h mean O<sub>3</sub> concentrations at selected rural sites (indicated as 1, 3, 5, 6, 8, 12, 16, 23 - see Tab. 1) in the CR for the period under consideration is summarized by a histogram showing that about 70 % of 1-h mean O3 concentrations were above 40 ppb (Fig. 4). The AOT40 Forest trends for different altitudinal layers are presented in Fig. 5. The AOT40 values for Czech rural sites clearly show that the yearto-year variability was considerable. The critical level of 5 ppm h (UN/ECE 2004) was exceeded every year at all Czech rural sites. In years with abundant O<sub>3</sub>, the critical level was exceeded 5-7.5 times (2003). The highest AOT40 values were recorded at the Sous site (39 ppm h in 1994), Prebuz and



**Tab. 4** - Average day of the year (DOY) when the critical level of 5 ppm h was exceeded in the 1994-2008 growing seasons at selected rural sites.

Veer	Site ID									
1 cai	1	3	5	6	8	12	16	23	Average	
1994	-	-	5.5	-	-	-	16.5	-	10.5	
1995	-	-	23.4	-	12.5	6.5	24.5	-	8.5	
1996	21.4	17.4	-	27.4	21.4	26.4	2.5	28.4	26.4	
1997	12.5	12.6.	14.5	29.5	17.5	15.5	16.5	19.5	18.5	
1998	10.5	25.5	9.5	-	8.5	21.6	12.5	10.5	20.5	
1999	30.4	22.5	8.5	18.5	17.5	13.5	19.5	28.5	15.5	
2000	7.5	30.4	4.5	6.5	7.5	5.5	7.5	7.5	5.5	
2001	20.5	14.5	24.5	23.5	25.5	25.5	9.6	23.5	27.5	
2002	9.5	7.5	14.5	14.5	12.5	17.5	17.5	21.5	14.5	
2003	29.4	26.4	6.5	6.5	4.5	6.5	5.5	6.5	5.5	
2004	26.5	31.5	27.5	31.5	1.6	13.5	7.6	30.5	28.5	
2005	12.5	-	8.5	16.5	5.5	3.5	16.5	17.5	9.5	
2006	18.4.	6.5	8.5	12.5	7.5	5.5	10.5	10.5	8.5	
2007	14.5	14.5	19.5	5.5	13.5	5.5	11.5	13.5	10.5	
2008	17.5	13.5	3.6	21.5	14.5	16.5	25.5	30.5	22.5	
Average	8.5	13.5	14.5	14.5	11.5	13.5	16.5	16.5	13.5	

Krkonose-Rychory sites (38 ppm  $\cdot$ h in 1994), and Bily Kriz (37.7 ppm  $\cdot$ h in 1995) in the North. Apart from these mountain sites, fairly high AOT40 was recorded also in lower altitude at the Mikulov site (34 ppm  $\cdot$ h in 2003).

The annual trend of AOT40 differed depending on the meteorological conditions. The critical level of 5 ppm h was usually reached in the beginning of the growing season (Tab. 4), with few exceptions. As an example, we present the accumulation of AOT40 over the growing seasons 2003-2008 at two sites: a mountain site (3. Krkonose-Rychory) and a rural site situated at a medium altitude (15. Kostelni Myslova - Fig. 6). Rapid AOT40 rise was recorded in 2003, when the critical level of 5 ppm h at the Krkonose-Rychory site was reached as early as before the end of April and at the Kostelni Myslova site in the beginning of May. Ne-



Fig. 6 - Annual trend of AOT40 at Krkonoše-Rýchory (1001 m a.s.l.) and Kostelní Myslová (569 m a.s.l.), 2003-2008 growing seasons.

vertheless, even in the years with meteorological situations not conducive to  $O_3$  formation, the critical level of 5 ppm h was usually reached during May.

Fig. 7 shows the spatial distribution of AOT40 in 2003 when the exceedances of 5 ppm h were the highest ever recorded. The critical level was exceeded more than 6 times over one third of the Czech forested area. The highest values were recorded over the border mountains - the Krkonose, the western part of the Krusne hory Mts., the Cesky les, the Sumava, and also inland over the Czech-Moravian Upplands (Ceskomoravska Vysocina) and the Brdy. Fig. 8 shows the spatial distribution of AOT40 in 2006 which belonged to the years abundant in O<sub>3</sub> (though lower as compared to the extreme year of 2003) as a consequence of a very warm and dry summer in Central Europe. In contrast to 2003, the relative share of forested area with AOT40 above 30 ppm h was < 3 %. The highest values were recorded in the South, in the Sumava and Novohradske hory Mts. Fig. 9 shows the year 2007 with even lower O3 levels. Still, 78 % of the Czech forested area experienced an exceedance of the critical level by a factor of 3-4.

### Discussion

The crucial factors for ambient O<sub>3</sub> concentrations are emission of precursors and prevailing meteorological conditions. Solar intensity is of particular importance. Hot sunny calm weather leads to high O3 concentrations. High temperature, high solar intensity, low wind speed, low atmospheric humidity and absence of precipitation are factors generally considered as favorable for photochemical O<sub>3</sub> formation (Seinfeldt & Pandis 1998). While anthropogenic O<sub>3</sub> precursor emissions in Europe have decreased (EEA 2007), the air temperature tends to increase. Observational evidence from all continents shows that many natural systems are being affected by regional climate changes, particularly temperature increases (IPCC 2007). For CR, Tolasz et al. (2007) reported a statistically significant temperature increase for the period 1961-2000: annual average temperature increases by 0.028 °C year-1 (R<sup>2</sup> = 0.195) and growing season average temperature increases by 0.025 °C year<sup>-1</sup> ( $R^2 = 0.151$ ). A similar trend is still observable and we can summarize that the average temperature in the CR increases by 0.3 °C per decade in the last 50 years (http://www.chmi.cz).

Ambient O<sub>3</sub> is a regional phenomenon and for its formation the emissions from broader regions are of importance. According to European Environmental Agency (EEA), emissions of the main ambient O<sub>3</sub> precursor pollutants have decreased significantly across the EEA region between 1990 and 2009 as follows: NO<sub>x</sub> by 44 %, non-methane VOCs by 55 %, CO by 62 % and CH<sub>4</sub> by 27 % (EEA 2010). These estimates correspond with the trend in the CR, though the O<sub>3</sub> precursor emission decrease in the CR was even more pronounced as compared to the EEA region. Though the uncertainties associated with estimated emissions are relatively high, accounting for about 50 % for VOCs and CH<sub>4</sub> and 30 % for NO<sub>x</sub> (De Leeuw 2002), the progress in reducing emissions is obvious. We have to keep in mind, however, that O<sub>3</sub> formation changes under differing NO<sub>x</sub> and VOC regimes (Seinfeldt & Pandis 1998), and tropospheric ozone-forming potential differs for individual precursor gases (De Leeuw 2002).

There is a discrepancy between the substantial cuts in  $O_3$  precursor emissions and observed non-decreasing annual average  $O_3$ concentrations in Europe. Reasons include increasing inter-continental transport of  $O_3$ and its precursors in the northern hemisphere, climate change, biogenic non-methane VOC emission, which are difficult to quantify, and natural fires (EEA 2010).

An inherent property of AOT40 as a cumulative index is that it is very sensitive to the quality of input data. For calculating AOT40Forest, O<sub>3</sub> concentrations were measured in real time, thoroughly checked and considered as highly variable; the calculated index, however, is not robust (Sofiev & Tuovinen 2001) and is likely to be burdened by high uncertainty. Consideration of the spatial scale is a crucial issue for air pollution mapping, as stressed by Diem (2003). A spatial resolution of 1 x 1 km, as used in our mapping, is detailed enough, provides consistent results, and is considered appropriate for a country-scale mapping (Gottardini et al. 2010). The relative uncertainty of the AOT40Forest maps analyzed by cross-validation and expressed by the root mean square error (RMSE) was about 20 % as shown earlier by Hůnová et al. (2012) in a comparison of 11 different interpolation approaches for ambient O<sub>3</sub> mapping. The relative uncertainty of AOT40Forest maps is worse when compared to maps of seasonal mean O<sub>3</sub> concentrations, but it is still acceptable

A similar approach for  $O_3$  phytotoxic potential assessment based on  $O_3$  concentra-



tions measured in real time and interpolation of calculated AOT40Forest values was used for United Kingdom by Coyle et al. (2002), and for EU by Horálek et al. (2007, 2009). Different interpolation techniques were used in these studies, but approach and scale were similar. For Italy, where monitoring network does not adequately cover all the territory, De Marco (2009) applied an integrated assessment model (RAINS-Italy) for developing a map of AOT40Forest.

The highest  $O_3$  concentrations in Europe are observed in the Mediterranean countries (EEA 2010). Nevertheless,  $O_3$  concentrations measured throughout central Europe (Matyssek et al. 2007) including the CR are also very high. The AOT40Forest values that we calculated for some Czech sites are comparable to values reported for Italy (Paoletti 2006).

Our analysis of real-time O3 data recorded during 1994-2008 within the framework of the Czech national ambient air quality monitoring network shows that O<sub>3</sub> exposure over the Czech forested areas still remains fairly high and varies considerably in time and space. We assume that in the Czech rural regions, where average hourly O<sub>3</sub> concentrations during the growing season are generally significantly higher than 40 ppb (Hůnová 2003, Hůnová et al. 2003) and, in particular, under optimal nutrient regime and water availability, the AOT40 exposure index provides a reasonable estimation of the risk areas. This assumption would not apply for the year 2003, an extreme year regarding meteorology (Luterbacher et al. 2004), when the high O<sub>3</sub> concentrations were coupled with extreme drought affecting stomatal conductance.

As stressed earlier by many authors, it is necessary to observe plant effects to give biological significance and meaning to O<sub>3</sub> standards (Manning 2003). Visible ozone injury is assessed regularly at selected plots within the forest condition monitoring in the CR by Forestry and Game Management Research Institute (Bohacova et al. 2010). Despite the high O<sub>3</sub> levels measured over the CR, no serious damage to vegetation attributable to O<sub>3</sub> has been reported so far. In a case study carried out in the Jizerske hory Mts. in 2006 and 2007 at five sites situated in the altitudes 900-1000 m a.s.l., the leaves of 22 plant species were assessed for ozonelike visible symptoms according to UN/ECE (2004). Though injury was found, the extent of visible symptoms was much less than assumed considering the recorded O<sub>3</sub> exposure (Matousková et al. 2010). Moreover, after verification of symptoms by the Ozone Validation Centre for Central Europe (Günthardt-Goerg & Menard 2008), visible injury was confirmed as O<sub>3</sub>-induced on the leaves of only two species, Fagus sylvatica and Rubus idaeus (Hůnová et al. 2011). This is

in agreement with earlier reports from many other authors (e.g., Ferretti et al. 2007, Matyssek et al. 2007, Waldner et al. 2007, Baumgarten et al. 2009, Bussotti & Ferretti 2009), that the  $O_3$  exposure is rather inconsistent with observed ozone injury. Moreover, the assumption that higher ambient O<sub>3</sub> exposure results in higher contents of malondialdehyde (MDA) as a product of lipid peroxidation in Picea abies needles was not supported by a study in real forest stands of three Czech mountain areas - the Krkonose, Krusne hory and Jizerske hory Mts. in 1994-2006 (Hůnová et al. 2010). In contrast, Srámek et al. (2007) indicated an impact of  $O_3$  on beech (*Fagus sylvatica*) as assessed by the quality and quantity of epicuticular waxes and content of MDA. Furthermore, Zapletal et al. (2011) reported that ambient O<sub>3</sub> reduces net ecosystem production in a Norway spruce (Picea abies) stand in the CR, though spruce as most of the conifers is considered as a relatively ozone-tolerant species (UN/ECE 2004). Thus, so-far reported impacts of O<sub>3</sub> on vegetation in the CR are equivocal and further effort is needed to clarify the issue and translate O<sub>3</sub> exposure to biological impacts on CR forests.

# Conclusions

Our analysis of real-time ambient  $O_3$  measurements at Czech rural sites recorded over the last 15 years shows that  $O_3$  exposure over the Czech forested areas still remains fairly high and varies considerably in time and space. All sites consistently exceeded the critical level of 5 ppm h AOT40Forest since 1994, with peak values reaching 38-39 ppm h at few sites in different years. Existing studies on ambient  $O_3$  biological effects on Czech forests are equivocal and more effort is needed to explore the  $O_3$  impacts. Regarding environmental protection the effort should be focused on the most sensitive forest species.

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