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Observation-based trends in ambient ozone in the Czech Republic over the past two decades

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ABSTRACT

We present the trends in ambient ozone concentrations based on high quality data measured continuously at 26 long-term monitoring sites (9 urban, 17 rural including 10 mountain stations) in the Czech Republic in 1994–2015. We considered annual and summer medians, the 10^{th} and 98^{th} percentiles, maximum daily 8-h running mean concentrations and exposure index AOT40F. For all indicators taken into account except for the 10^{th} percentile, our results showed a similar pattern with significant decreasing trends for about one half of the examined sites. We obtained similar results for all types of sites. The most pronounced decrease in O_3 concentrations was recorded at mountain sites. Namely, at the Šerlich mountain site, with an overall decrease per year in annual median by 0.43 ppb, summer median by 1.17 ppb, maximal daily 8-h average by 0.45 ppb, the 10^{th} percentile by 0.62 ppb. The peak concentrations indicated by the 98^{th} percentile and AOT40F decreased most at urban site České Budějovice by 0.75 ppb and 0.84 ppb h per year, respectively. For sites exhibiting significant decreasing trends, an overall decrease per year in annual median was 0.22 ppb, in summer median 0.41 ppb, in the 10^{th} percentile 0.23 ppb, in the 98^{th} percentile 0.53 ppb, and in AOT40F 0.51 ppb h. A significant increasing trend was detected only in the 10^{th} percentile at just three sites, with the highest increase of 0.19 ppb per year recorded at the rural site Sněžník. Moreover, a consistent decrease in limit value exceedances was detected, with by far the highest violation recorded in the meteorologically exceptional year of 2003.

Out of the 26 sites under review, seven have not recorded a significant decreasing trend in O_3 in any of the considered statistics. The lack of trends in O_3 at these seven sites is likely associated with changing time patterns in local NO and NO_2 emissions: in particular, with the increasing ratio in NO_2/NO_x. There is an obvious geographical pattern in recorded O_3 trends: most of the sites with no trend detected are situated in the Northwestern region of the CR with numerous energy-producing large emission sources, partly denitrified recently. Our results clearly indicated that, for O_3 decrease, the ratio between individual NO and NO_2 forms is critical, and that a simultaneous significant decrease in both NO and NO_2 concentrations is not a sufficient prerequisite. Apart from changes in car fleet in urban areas or near motorways, this factor might be of particular relevance in formerly highly polluted areas, where emissions from large power plants recently substantially decreased.

1. Introduction

Ground-level ozone (O_3), a key component of ambient air (Monks et al., 2015), has been extensively studied in recent decades from different aspects: with regard to its atmospheric chemistry, in context of climate change (Isaksen, 2003; Simpson et al., 2014), and with respect to its impacts (e.g. Anenberg et al., 2010; Paoletti, 2007). Being a secondary pollutant, O_3 is formed from precursors during complex photochemical reactions, in highly non-linear O_3-NO_x-VOC chemistry, and it is extremely difficult to reduce its ambient concentrations (Seinfeld and Pandis, 1998; Finlayson-Pitts and Pitts, 2000). Despite several decades of control in North America, Europe and Japan, the North Hemispheric background O_3 concentrations are increasing (Derwent et al., 2004), mostly due to the anthropogenic activities of growing populations of the East-Asian region, China in particular (Ma et al., 2016; Granier et al., 2011; Wang et al., 2017). Thus, O_3 remains a serious air pollution problem (Royal Society, 2008; Monks et al., 2015), and represents a major threat both for human health and ecosystems (EEA, 2016). In Europe, the critical levels for O_3 are permanently exceeded over vast areas (EEA, 2016). With respect to its importance, it is no wonder that numerous studies presenting trends in O_3 ambient levels in different regions have been published (e.g. Vingarzan, 2004; Sicard et al., 2013; Cooper et al., 2014).

In the Czech Republic (CR), the country infamous for serious air

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pollution problems in the past (Moldan and Schnoor, 1992), in particular due to enormous SO_2 emissions in the 1970s and 1980s, O_3 as an ambient air pollution issue emerged as late as in the 1990s. Monitoring of O_3 concentrations has been done since 1993. At first, monitoring occurred at just a few sites, increasing over time to some 50 sites currently in operation, including both rural and urban stations, covering the country (CHMU, 2016). O_3 levels measured so far show that the highest levels are recorded in areas considered as "clean" regarding other ambient air pollutants, such as PM, SO_2, and NO_x (Hůnová, 2003). O_3 exposures in the CR are relatively high (Hůnová et al., 2003; Hůnová and Schreiberová, 2012), and may result in negative endpoints both regarding human health (Hůnová et al., 2013) and vegetation (Hůnová et al., 2011; Vlasáková-Matoušková and Hůnová, 2015).

Ozone formation is highly meteorology dependent (Finlayson-Pitts and Pitts, 2000) and thus its levels vary considerably from year to year depending on meteorological conditions. Nevertheless, such a long series of O_3 concentrations resulting from continuous monitoring allow for a sound trend analysis. The aim of this paper was to analyse ambient O_3 for trends at Czech long-term monitoring sites over 1995–2015. Additionally, we inspected the time changes in ambient NO and NO_2 levels, and in concentration ratio in NO_2/NO_x (on an annual basis), as these are strongly interlinked with O_3 concentrations.

2. Methods

2.1. Sites and period under review

For our analysis we used 26 out of 54 existing Czech sites measuring O_3 (Table 1, Fig. 1). These are included in the nation-wide ambient air quality monitoring network, and operated by the Czech Hydrometeorological Institute. Apart from O_3 levels, they also monitor other air pollutants of interest, in particular in view of compliance with the legal limit values (CHMU, 2016). All these sites are included in the EU database AIRBASE.

Table 1
Measuring sites selected for our trend analysis (listed according to increasing altitude).

Site	Region	Classification	Altitude [m a.s.l.]
Urban sites (9)			
Ostrava-Fifejdy	Moravskoslezský	B/U/R	220
Karviná	Moravskoslezský	B/U/R	238
Hradec Králové-observatoř	Královéhradecký	B/S/R	276
Praha 4-Libuš	Hl. m. Praha	B/S/R	301
Plzeň-Doubravka	Plzeňský	B/S/A	348
Ústí n.L.-Kočkov	Ústecký	B/S/RN	367
České Budějovice	Jihočeský	B/U/R	383
Sokolov	Karlovarský	B/S/R	476
Prachatice	Jihočeský	B/S/R	583
Rural sites (7)			
Mikulov-Sedlec	Jihomoravský	B/R/A-REG	245
Tušimice	Ústecký	B/R/IA-NCI	322
Ondřejov	Středočeský	B/R/N-REG	514
Košetice	Vysočina	B/R/N-REG	535
Kostelní Myslová	Vysočina	B/R/A-NCI	569
Sněžník	Ústecký	B/R/N-REG	590
Svratouch	Pardubický	B/R/N-REG	735
Mountain sites (10)			
Jeseník-lázně	Olomoucký	B/R/N-NCI	625
Přimda	Plzeňský	B/R/N-REG	740
Souš	Liberecký	B/R/N-REG	771
Hojná Voda	Jihočeský	B/R/N-REG	818
Rudolice v Horách	Ústecký	B/R/N-REG	840
Bílý Kříž	Moravskoslezský	B/R/N-REG	890
Přebuz	Karlovarský	B/R/AN-REG	904
Krkonoše-Rýchory	Královéhradecký	B/R/N-REG	1001
Šerlich	Královéhradecký	B/R/N-REG	1011
Churáňov	Jihočeský	B/R/N-REG	1118

Note.: Sites are classified according to EC Decision EoI 97/101/EC (EC, 1997):
B/S/R – background/suburban/residential.
B/U/R – background/urban/residential.
B/S/RN - background/suburban/residential, natural.
B/R/A - background/rural/agricultural/.
B/R/N-REG - background/rural/natural-regional.

The selection criteria of examined sites were the length and completeness of their records. The time periods over which the O_3 trends for individual stations were analysed slightly differed between sites to make use of all data available. The time span between the beginnings of measurements at individual sites is between 1994 and 1997. That means that the longest periods available were 22-year series (for eight sites), and the shortest, 19-year series (for three sites), for details, see Table 2. Nevertheless, working with an average over specific type of sites (i.e. rural, mountain, urban), we considered the time span of 1995–2015, to be able to include most sites into the analysis.

With respect to geographical coverage, the sites under review cover unevenly the entire Czech Republic, representing both rural and urban areas. Regarding the site classification, we analysed 9 urban and 17 non-urban sites: from the rural, 10 sites were situated in mountain areas. Unlike the urban locations, the rural sites (EC, 1997), with no important emission sources nearby, are expected to represent considerable areas, including mostly tens to hundreds of kms, as they are influenced exclusively by long-range or regional air pollution transport. Unlike EoI classification of measuring sites (EC, 1997), from the rural sites we excluded mountain sites as a specific subgroup, and considered these two separately, due to the fact that O_3 is highly dependent on global solar radiation, which increases with increasing altitude, and thus O_3 levels in mountains are expected to be higher. Mountain sites, however, were not classified solely based on altitude. We also considered the macrosetting of the stations. For example, Svratouch situated on a solitary hill at an altitude of 735 m a.s.l. was included into rural sites. By contrast, Jeseník-lázně, situated in the Jeseníky Mts., at an altitude of 625 m a.s.l., was included into mountain sites, although its altitude is lower than Svratouch.

2.2. Ozone data

We used O_3 concentrations measured continuously, year-round, within the national ambient air quality monitoring network by UV absorbance, the EC reference method (EC, 2008). The ozone analysers used were the Thermo Environmental Instruments (TEI), M49. The samplers were set up within the breathing zone, some 2 m above the ground. The equipment and procedures did not change over the time series. Standard quality assurance/quality control (QA/QC) procedures were applied (EC, 2008). The input data were 1 h mean concentrations, the basic values stored in the nation-wide ambient air pollution database ISKO (CHMU, 2016).

2.3. NO_x data

As well as ozone data analysis, we checked the time changes in NO, NO_2 and NO_x ambient levels as these are strongly interlinked with O_3 concentrations. In particular, we looked at the concentration ratio in NO_2/NO_x on an annual basis. Nevertheless, we have not gone into more detail, as it was beyond the scope of this paper.

We used NO, NO_2 and NO_x data measured continuously, year-round, within the national ambient air quality monitoring network by chemiluminiscence, the EC reference method (EC, 2008). The analysers used were Thermo Environmental Instruments (TEI), M42 until March 2015, and Teledyne Advanced Pollution Instruments (TAPI), T200 since March 2015. The setting, QA/QC and storage were the same as mentioned above for O_3.

2.4. Statistical analysis

We studied six characteristics of O_3 concentrations reflecting low, mean and peak levels. We considered annual and summer (April-