# CONCENTRATION OF THE SELECTED AIR POLLUTANTS AT BÍLÝ KŘÍŽ, THE CZECH REPUBLIC, FOR 1994 TO 1999

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# ABSTRACT

Air pollution related problems, such as acidification, air quality and ground-level ozone, are recognized as some of the important problems facing the people of the Czech Republic, whose health is damaged by impacts of air pollutants. Also the impacts of these air pollutants on human health, forests and vegetation are investigated. This paper presents evaluation of air pollution caused by sulfur dioxide, nitrogen oxides, particulate matter and ozone, based on the readings of measuring site at Bílý Kříž for the six-year period 1994-1999. The evaluation of the status of the ambient air pollution is based on air pollution limit values as specified by the new European Directives. Concern has been expressed about the possible effects of ozone, nitrogen oxides, sulfur dioxide and particulate matter exposures on forested areas, materials and human health. The main objective of this study is to examine the evolution observed in the concentrations of these air pollutants considered. The reduction is observed for winter air pollutants as sulfur dioxide and particulate matter from 1996 to 1999. In contrary, the ozone concentrations increase considerably for the same period.

# INTRODUCTION

Air pollution in the Czech Republic, especially in the forested stations, as Bilý Kříž, has been the subject of many studies from national and international research groups during the past years (Rožnovský and Blažek, 1996; Černý, 1999; Knotková and Kreislová, 1999; Manczyk, 1999; Zapletal, 1999; Hůnová *et al.*, 2000). The world is making progress to reduce some or all air pollutants and the researchers are interested by their impacts on human health and others ecosystems (Fally *et al.*, 1995; Andreani-Aksoyoglu, 1996; Bisson *et al.*, 1997; EEA, 1997; APPA, 1998; Beck *et al.*, 1997; Lefohn and Shadwick, 1998; Molnárová and Mindáš, 1998; Kalabokas *et al.*, 1999). These harmful substances are ozone, nitrogen oxides, sulfur dioxide, particulate matter, carbon monoxide, carbon dioxide, methane and so on.

It is important to know the level accepted by human, vegetation and animals. For all efforts to limit air pollutants in the atmosphere, their concentrations remain high into the cities all around the world. The ambient pollution is due to the use of fossil combustible, increase of traffic roads, industrial processes. NO<sub>x</sub> is a precursor in the formation of tropospheric ozone in photochemical reaction, which is also a greenhouse gas and together with SO<sub>2</sub> causes the phenomenon of acid rain.

Pollutant	Formula	Air pollution standards (µg m <sup>-3</sup> )			
		IHr	IHd	IH8h	IH1h
Sulfur dioxide	SO <sub>2</sub>	20	125		
Particulate matter	PM <sub>10</sub>	40	50		
Nitrogen oxides	NO <sub>x</sub>	30	40*		
Ozone	O <sub>3</sub>	40		120	180

Table 1. Air pollution standards for European Union

\* for NO<sub>2</sub>

IHr – average annual pollutant concentration

IHd – average daily pollutant concentration

IH8h - average eight-hour pollutant concentration

IH1h – average one-hour pollutant concentration

Increased tropospheric ozone concentrations are currently a matter of large concern, as the key environmental problems. Recent studies have been shown that elevated ozone concentrations may affect strongly human health, vegetation and materials (EEA, 1997; Beck et al., 1998; Závodská et al., 1998; Kalabokas et al., 1999; Vecchi and Valli, 1999; Hůnová et al., 2000; Jackson et al., 2000; Molnárová, 2000). The mechanism for ozone production is the oxidation of anthropogenic and biogenic volatile organic compounds (VOCs), methane ( $CH_{4}$ ), nitrogen oxides ( $NO_{x}$ ) and carbon monoxide (CO) (Rožnovský and Blážek, 1999; Závodská et al., 1998; Hůnová et al., 2000; Jackson et al., 2000; Molnárová, 2000). Ambient ozone concentration, a component of the ozone exposure of plants, is the parameter used for the ozone critical levels. In Europe, the highest ozone concentrations occur in summer, under stable high-pressure systems with clear skies (Fally et al., 1995; Závodská et al., 1998; Kalabokas et al., 1999; Vecchi and Valli, 1999; Hůnová et al., 2000). According to Badot et al. (1988), EEA (1997), Beck et al. (1998) and Manczyk (1999), in some polluted areas in the Czech Republic, Germany and Poland the forest is heavily damaged.

#### DATA SOURCES

Czech Hydrometeorological Institute network: Systematic measurements of air pollutant concentrations are in operation since the seventeenth century. All stations implanted in the Czech Republic measure concentrations of sulfur dioxide, nitrogen oxides, ozone, suspended particulate matter, carbon monoxide, carbon dioxide, volatile persistent organic pollutants, heavy metals, aromatic hydrocarbons, etc. Since 1981, on the basis of an agreement between the Ministry of Forests and Water Management and the Ministry of Health, the Czech Meteorological Institute was entrusted with overall processing and the National Health Institute was designated as the collective workplace for the health sector.

### **EVALUATED STATION**

The considered station for this study is Bílý Kříž. It is located at an altitude of 890 m. at 49°30'17" northern latitude and 18°32'28" eastern longitude in the region of the Beskids. The air pollution measurement methods are the measuring methods in the network of automated monitoring stations (AMS). The region of Beskids, especially Bílý Kříž, is about 30 to 40 km from important emission sources in the regions of Karvina and Ostrava. SO<sub>2</sub> concentration is measured by the ultraviolet fluorescence method, model SA 631. The radiometric method is used for PM<sub>10</sub> concentration, model DUB 32. The concentration of  $NO_x$  is measured by a chemiluminescence's analyzer, model NA 621. The ozone concentration measurement is based on ultraviolet absorption photometry, by UV analyzer by Te Instruments, model 49.

Assessment of the status of the air pollution is



*Figure 1.* Annual concentration of SO<sub>2</sub>, with the annual limit of 20 µg m<sup>-3</sup>, Bílý Kříž, 1994-1999



*Figure 3.* Annual concentration of NO<sub>x</sub>, with the annual limit of 30 μg m<sup>-3</sup>, Bílý Kříž, 1994-1999

based on air pollution standards as specified by new European Directive, published on 22 April 1999 (1999/30/EC). The levels of pollution are characterized by daily, monthly, annual, winter and summer average according to the new directive of air pollution standards in Europe.

#### EVOLUTION OF THE ANNUAL AVERAGES

The evolution of annual averages of  $SO_2$ ,  $PM_{10}$ ,  $NO_x$  and  $O_3$  between 1994 and 1999 are given in Figs 1-4. The limits of air pollutant concentrations recommended by the European Union are plotted as horizontal lines.

# **SO**<sub>2</sub>

Sulfur dioxide is formed when fuel containing sulfur is burned and during metal smelting and other



Figure 2. Annual concentration of PM<sub>10</sub>, with the annual limit of 20 μg m<sup>-3</sup>, Bílý Kříž, 1994-1999



Figure 4. Annual concentration of O<sub>3</sub>, with the annual limit of 40 μg m<sup>-3</sup>, Bílý Kříž, 1994-1999

industrial processes (Blažej et al., 1981; Tolgyessy et al., 1984; Jonaš, 1985; Burki and Keller, 1994). High concentrations of SO<sub>2</sub> can cause wheezing, shortness of breath and cardiovascular diseases (Fally et al., 1995; EEA, 1997; APPA, 1998). But together, SO<sub>2</sub> and NO<sub>x</sub> are major precursors to acidic deposition, such as acid rain, and accelerate corrosion of buildings and monuments. The daily and annual data are considered for determination of air pollution level for protection of human health and of vegetation respectively. The SO<sub>2</sub> level is continuously decreasing due to the improved quality of the fossil fuels. The most critical year was 1996, with the concentration of 20.4  $\mu$ g m<sup>-3</sup> (Fig. 1). Comparing to other stations, Bílý Kříž presents low levels of pollution caused by sulfur dioxide (Fig. 5). Significant decrease of this



Figure 5. Annual concentration at selected stations for 1996-1998, SO<sub>2</sub>

type of pollution was confirmed in 1999. Annual values fluctuated between 7.5 and  $20.4 \,\mu g \, \text{m}^{-3}$ , as shown in Fig. 1. The decrease in sulfur dioxide concentrations was caused by favorable meteorological and dispersion conditions, especially in winter, by the decrease in emissions from the main sources, and also because of the station proximity to the sources of emission, the elimination of the oldest equipment in various power plants and the desulfurisation by many industries (Manczyk, 1999).

#### PM<sub>10</sub>

Particulate matter is the general term used for a mixture of solid particles and liquid droplets found in the air. The  $PM_{10}$ , the coarse particles, are generally emitted from sources, such as vehicles traveling on unpaved roads (Blažej *et al.*, 1981; Tolgyessy *et al.*, 1984). In addition to health problems (respiratory problems, asthma,

decreased lung function, premature death), PM<sub>10</sub> is the major cause of reduced visibility and can damage paints and building materials. Annual PM<sub>10</sub> values clearly remain above the limit of 20  $\mu$ g m<sup>-3</sup> from 1994 to 1996 (Fig. 2). Compared to other stations (Fig. 6), Bílý Kříž station registered low particulate matter concentrations for 1996-1999. Figure 2 show that in 1998 the  $PM_{10}$  pollution decreased significantly, at 16.3  $\mu$ g m<sup>-3</sup> compared to 32.9  $\mu$ g m<sup>-3</sup> in 1994. The annual concentration of PM<sub>10</sub> goes down from 1994 to 1999 because of the decrease in the emission of this pollutant from the sources. This tendency was observed around the Czech Republic for the all measured period, according to Ministry of Environment (Petružela et al., 1999). Also the drop of temperature influenced the decrease in PM<sub>10</sub> concentrations at the station of Bílý Kříž like around the Czech Republic for cold halfyears 1997-98 and 1998-99 (Petružela et al., 1999).



*Figure 6.* Annual concentration at selected stations for 1996-1998,  $PM_{10}$ 



Figure 7. Annual concentration at selected stations for 1996-1998, NO<sub>x</sub>

# NO<sub>x</sub>

Nitrogen oxides  $(NO_x)$ , the term used to describe the sum of NO and NO<sub>2</sub>, play a major role in the formation of tropospheric ozone and contribute to the formation of acid rain (EEA, 1997; APPA, 1998). Long-term exposures (e.g. one year) to nitrogen oxides concentrations may damage forestry and vegetation (Fally et al., 1995; EEA, 1997). Nitrogen oxides are formed at high temperature combustion processes, mainly from petrol and diesel vehicles (Blažej et al., 1981; Tolgyessy et al., 1984; Burki and Keller, 1994). Pollution levels above  $15 \,\mu g \, m^{-3}$  were not recorded at Bílý Kříž station. But in comparison with other stations, Bílý Kříž registered the lowest annual values, ranging between 7.8 and 11.1  $\mu$ g m<sup>-3</sup> for 1994-1999 (Figs. 3 and 7). The annual limit of 30  $\mu$ g m<sup>-3</sup> was not exceeded in any considered period at Bílý Kříž and a slight decrease was observed in nitrogen oxides concentrations in comparison of 1999 with 1994. The moderated decrease in nitrogen oxides concentrations was partly due to favorable meteorological and dispersion conditions, mainly in winter period and to a decrease of NO<sub>x</sub> emission from stationary sources in comparison to previous years. This drop is also due to the amelioration in the equipment in power plants (Manczyk, 1999).

# **0**<sub>3</sub>

Ozone is classified as a secondary pollutant in the troposphere. It is formed in the air as a result of chemical reactions of  $NO_x$  and VOCs, its precursors, under the influence of solar radiation (Brasseur, 1982; Lefohn *et al.*, 1994; Rožnovský

and Blažek, 1996; EEA, 1997; APPA, 1998; Závodská et al., 1998; Schweitzer, 1999). Groundlevel ozone is a gas on which attention has becoming increasingly focused in the context of its negative impacts on all types of receptors, human, animal, vegetation, monuments and buildings (Jonaš, 1985; Badot et al., 1988; Lefohn and Lucier, 1991; Mage and Zali, 1992; Bisson et al., 1997; EEA, 1997; APPA, 1998; Beck et al., 1998; Černý, 1999; Musselman and Massman, 1999; Vecchi and Valli, 1999; Schweitzer, 1999; Hůnová et al., 2000, Jackson et al., 2000; Molnárová, 2000). Short-term (one hour) and prolonged (8 hours) exposures to ambient ozone have been linked to a number of health effects of concern. The longterm (e.g. 3 months, 6 months, one year) ozone exposure affects vegetation and ecosystems, reduces the plant growth and increases the plant susceptibility to diseases. The current ambient air pollution limit value for ozone is  $120.0 \,\mu g \, m^{-3}$ , the eight-hour mean. The annual limit for the protection of vegetation is 40  $\mu$ g m<sup>-3</sup>. For 1995 to 1999 the critical annual value was  $88.02 \,\mu \text{g m}^{-3}$  in 1995 and the lowest one was 68.46  $\mu$ g m<sup>-3</sup> in 1996 (Tshiamala and Rožnovský, 2000a). Figure 4 shows that from 1997 to 1999 the annual value fluctuated between 76.25 to 78.44  $\mu g$  m<sup>-3</sup> (Tshiamala and Rožnovský, 2000b). The meteorological conditions were favorable for ozone formation in all considered period at Bílý Kříž, with higher frequency of limit values exceedences in the summer period according to Petružela et al. (1999) and Tshiamala and Rožnovský (2000b). The high ozone concentrations implicated that the forest ecosystem can be damaged. The





increase in 1998 was observed through the Czech Republic for the same time (Petružela *et al.*, 1999) and through Europe (APPA, 1998; Molnárová and Mindáš, 1999; Závodská *et al.*, 1998). The exposure index called AOT40 (Accumulated Exposure Over a Threshold of  $80 \,\mu g \, m^{-3}$ ) is determined for 1996 to 1999 at the Bílý Kříž station (Tshiamala and Rožnovský, 2000b). The AOT40 for forest is calculated for daylight hours only during six months period from April to September (vegetation period) and three months period from May to July (summer period). Figures 8-9 present the increased tendency of AOT40 values at the station of Bílý Kříž and the exceedance of AOT40 limit for all measured period.

# SEASONAL VARIATIONS OF MONTHLY AVERAGES Winter variations

In winter, episodes may occur when pollutants generated by the burning of fossil fuel accumulate in the atmosphere. These pollutants are sulfur dioxide, nitrogen oxides and suspended particulate matter (Fally et al., 1995; Andreani-Aksoysglu, 1996) and cause the winter smog with dramatic effects on human health (Ramade, 1991; Andreani-Aksoyoglu, 1996). The winter concentrations of PM<sub>10</sub> are upper than the limit of  $20 \ \mu g \text{ m}^{-3}$  from 1994-95 to 1996-97 (Fig. 2). The most critical winter was 1995-1996 with the concentration of 31.1  $\mu$ g m<sup>-3</sup>. The concentrations for the winter period decreased twice since 1995-96. Winter values of NO<sub>x</sub> are under the annual limit of 30  $\mu$ g m<sup>-3</sup> for all the measured period at Bílý Kříž. They varied between 9.0 and  $13.4 \,\mu g \, m^{-3}$  and



Figure 9. AOT40 for vegetation with the limit of 6,000 μg m<sup>-3</sup>·h, Bílý Kříž, 1996-1999

slowly decreased to 1998-99 (Fig. 3). Winter concentrations of SO<sub>2</sub> are higher than the annual limit of 20.0  $\mu$ g m<sup>-3</sup> for 1994-95 to 1995-96 and began to decrease since 1995-96 to 1998-99. The highest concentration measured for 1995-96 was 28.6  $\mu$ g m<sup>-3</sup>, but the lowest was 11.4  $\mu$ g m<sup>-3</sup> for 1998-99 (Fig. 1) according to Petružela *et al.* (1999). This situation is viewed around the Czech Republic because of the more favorable meteorological and dispersion conditions in the last winter periods, in contrary to 1995-96.

#### Summer variations

Figure 8 shows that the AOT40 index from April to September for the protection of forests exceeded the critical AOT40 limit of  $20,000 \,\mu \text{g m}^{-3} \cdot \text{h}$  for 1996-99 period at the Bílý Kříž station. Figure 9 presents the increased tendency of AOT40 values in summer (May to July) at the station of Bílý Kříž for the protection of culture and vegetation. This index exposure was exceeded more than twice in 1996, until three times up in 1999, than the AOT40 limit of 6,000  $\mu$ g m<sup>-3</sup> · h for the protection of vegetation from May to July. From April to September, in 1996 the lowest values were registered, and June, August and May presented the highest concentrations in 1996, 1997, 1998 and 1999, respectively. August is characterized by the highest concentrations of ozone at the other stations in the Czech Republic for the same period (Černý, 1999; Zapletal, 1999; Tshiamala and Rožnovský, 2000a, b). Ozone episodes are registered during the summer as APPA (1998), Fally et al. (1995) and Kalabokas et al. (1999) emphasize.

#### CONCLUSION

Data presented for the six-year period showed that the primary air pollutants concentrations such as nitrogen oxides, sulfur dioxide and particulate matter decreased year by year. But the tropospheric ozone presently increased noticeably, especially from 1996 at the Bílý Kříž station. All critical indices for ozone were exceeded considerably for the studied period. Human health, material, forestry and vegetation were considerably damaged by air pollutants, especially from tropospheric ozone at the Bílý Kříž station, in the region of Beskids. The decrease of sulfur dioxide, nitrogen oxides and particulate matter at the Bílý Kříž station was caused by favorable meteorological condition during the selected period, by decrease of the pollutants emissions from their sources. This drop was registered because of the proximity of forested station from the sources emission; the elimination of old equipment helped in reducing air pollutants emissions by many industries and power plants. The forested station of Bílý Kříž is damaged by high ozone concentrations, especially in summer and for the growing season.

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