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## HISTORICAL CHANGES IN AIR POLLUTION IN THE TRI-BORDER REGION OF POLAND, CZECH REPUBLIC AND GERMANY

In this study, we show the trends in the concentration of SO<sub>2</sub> and particulate matter (PM) in two health resorts, located in the tri-border region of Poland, Germany and Czech Republic. We analyze the annual time series and the seasonal variability of PM concentration for the months of July and February over the period of 1996–2007. Additionally, in July 2006, we measured the mean 24-hour concentration of PM and the content of heavy metals (by EDXRF analysis). We prove that nowadays air pollution in this region has diminished to a large extent as compared to the 90s of the last century. In Cieplice, the local influence is still evident; while Czerniawa is exposed to a periodical advection of polluted air from regional sources.

### 1. INTRODUCTION

The location of Lower Silesia in the region with old and geologically diverse mountains makes it rich in various natural resources, like copper ores, coal, brown coal, etc. Moreover, this region is very famous for its health resorts offering mineral waters, whose presumed healing properties have been proved for ages. They are well-known not only in Poland, but also in many other European countries (especially in the adjacent ones) and each of these resorts is frequently visited by foreign tourists,

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also from such countries as Denmark, Belgium or the Netherlands. Hence, it is essential to preserve the clean and fresh environment in order to allow and support the economical, social and tourist development of the region.

In the 70's and 80's of the XX century, this region together with Saxony and Northern Bohemia (the so-called "Black Triangle") was considered to be one of the most polluted places in Europe. The high density of glass factories, non-ferrous smelters and power plants without efficient gas cleaning techniques, a widely open coal mining with large erosion area, and the domestic heating on the basis of brown coal briquettes affected doubtlessly the air composition in this area [1]. How has the air quality changed since that time? Monitoring data are important to investigate the trends in time series of air pollutants and to observe the effects of reconstructing the major emission sources.

The study was focused on the investigation of SO<sub>2</sub> and PM concentration changes in ambient air in health resorts located in the Polish part of the Black Triangle region after the closing down and modernization of some industrial plants.

One of the objectives of Clean Air for Europe Project (CAFE) is "to develop, collect and validate scientific information on the effects of air pollution" (including validation of emission inventories, air quality assessment). Our investigation fulfils this task as it provides a scientific input which may be vital for implementing CAFE in a specific region – health resorts. CAFE should be addressed particularly to these areas.

## 2. METHODS

Air quality data (NO<sub>2</sub>, O<sub>3</sub>, CO, SO<sub>2</sub> and PM<sub>10</sub>) were obtained from the Voivodship Inspectorate for Environment Protection in Wrocław. All of those data were quality-controlled in the data management system.

We have used the monitoring data from two stations in the region of the Jelenia Góra Valley. Two sites were selected to represent the health resort area: Cieplice (50°54'N, 15°44'E, 330 m asl) in an urbanized region (urban background – in a park) and Czerniawa (50°54'N, 15°20'E, 650 m asl) in a relatively remote place (rural background). The monitoring stations have been operating since 1996. Additionally, field campaigns were run in Czerniawa, Świeradów and Cieplice in July 2006, 2007 and in February 2007, 2008. 24-hour aerosol samples were collected on Nuclepore filters, the part of a stacked filter unit connected to a medium-volume pump (2.4 m<sup>3</sup>/h flow rate). Weight concentration was determined gravimetrically. The concentration of elements was measured with an Energy Dispersive X-ray Fluorescence (XRF) spectrometer Epsilon 5 (PANalytical, Amelo, the Netherlands).

Meteorological parameters (wind speed and direction, temperature, relative humidity, pressure and radiation) were measured in an automatic weather station installed near the sampling point.

Backward air trajectories were calculated using the NOAA HYSPLIT (Hybrid Single Particle Lagrangian Integrated Trajectory) model [2] to estimate the long-range transport of air masses.

### 3. RESULTS AND DISCUSSION

Since the early 1960s, the forest destruction in Eastern Europe has been of major concern due to the extent and severity of the damage observed. Huge areas of forest have been suffering a serious deterioration in their health conditions, presenting strong visual symptoms of ecological damage. High depositions of sulphur acting synergistically with other factors are believed to have been responsible for extensive deforestation of spruce monocultures, especially in the mountainous regions. Figure 1 illustrates historical sulphur dioxide emissions in Poland, the Czech Republic and Germany [1] together with the estimates for the years 2010 and 2020 [3].

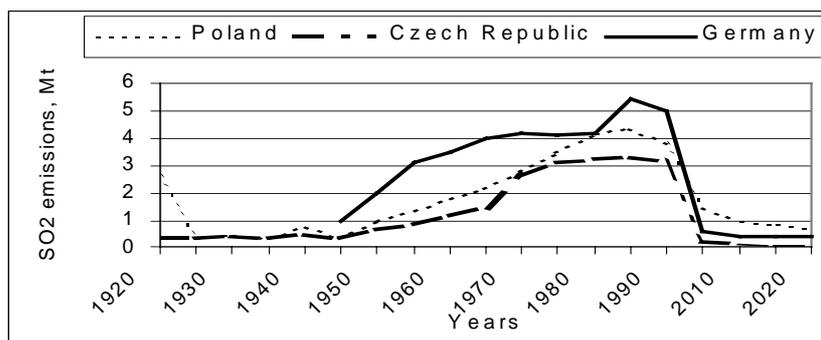


Fig. 1. Historical SO<sub>2</sub> emissions in Poland, the Czech Republic and Germany

Annual emissions of SO<sub>2</sub> in each country show, in general, an upward trend in the years 1945–1985, and a decrease from 1990. The sharp decrease of emission between 1989 and 1991 was caused by the collapse of the whole industrial structure in former East Germany, Czechoslovakia and Poland. A fast and efficient modernization of power plants was initiated. Many old factories and power plants, particularly in the former GDR, were closed [4]. In the following years (1992–1996), the emission reduction was mainly due to air-pollution control management, such as installation of flue gas desulphurisation equipment and changes in technological processes (bed-fluidized boilers). The emissions of SO<sub>2</sub> as well as of major air pollutants are intended for further decline if economic growth takes place over the period of 2000–2020 [3].

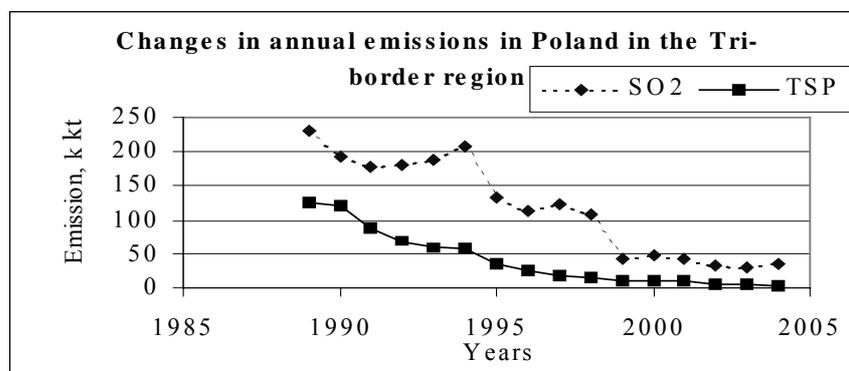


Fig. 2. Changes in annual SO<sub>2</sub> and TSP emissions in Poland in the tri-border region [5]

Figure 2 shows the changes in annual SO<sub>2</sub> and TSP emissions in Poland in the tri-border region [5]. A comparison of the data suggests a stronger decline in SO<sub>2</sub> than in TSP. The reduction of PM emission between 1989 and 1997 was relatively constant compared to that of SO<sub>2</sub>. The minor increase of SO<sub>2</sub> emissions was observed in 1994 and 1997. Long-term time series of pollutant concentrations offer the opportunity to follow the changes of their concentrations in ambient air over long time periods and to reproduce the emission trends. In order to compare the data, the annual mean values of PM concentrations in Czerniawa and Cieplice for the years 1996–2007 are shown in figure 3. It can be seen from this figure that the level of PM has decreased greatly. Nevertheless the pollution with PM is believed to be still an actual problem, particularly in urban sites.

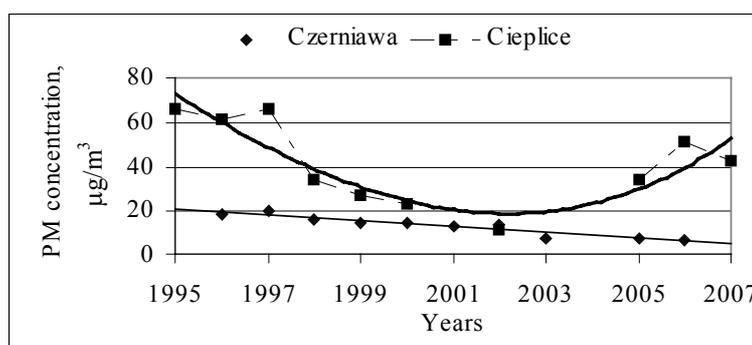


Fig. 3. Annual mean values of PM concentrations in Czerniawa and Cieplice for years 1996–2007

The seasonal variability of PM concentrations in urban and rural areas was investigated for the months of July and February over the period of 1996–2007 (figure 4).

At the beginning we wanted to discover whether or not the PM concentrations within July and February showed significant differences in the years 1996–2007. A Student's *t*-test for independent samples was used and the null hypothesis that the daily means of univariate sample population were equal was tested. Contrary to Cieplice, in Czerniawa no significant differences were found in the years 2004–2007. In Czerniawa, the concentrations of PM10 show a downward trend, i.e. from the daily mean value of  $16 \mu\text{g}/\text{m}^3$  in July 1997–1998 to the value of about  $8 \mu\text{g}/\text{m}^3$  for 2004–2007. In Cieplice, daily PM10 concentrations diminished in the years 1999–2000, then they remained constant till 2004 and started to rise since 2004, both in July and February.

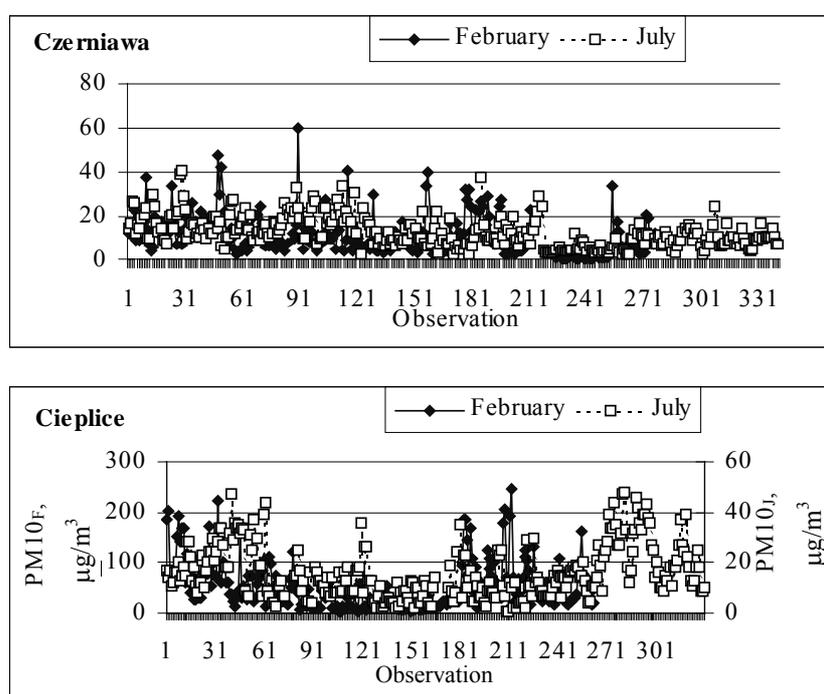


Fig. 4. Seasonal variability of PM concentrations in Czerniawa and Cieplice for months of July and February over period of 1996–2007

The overall results of our analysis indicate that the concentration of PM10 from regional sources has been decreasing since 1996, but local influences are still evident in the urban areas. The data shows that during both summer and winter PM10 concentration in Cieplice is higher than that in Czerniawa, and this is pronounced especially in winters. It was also interesting that PM10 concentrations in Czerniawa did not show any significant seasonal differences. The best conditions for longer residence time of PM in the atmosphere occur during warm and dry seasons. The level of

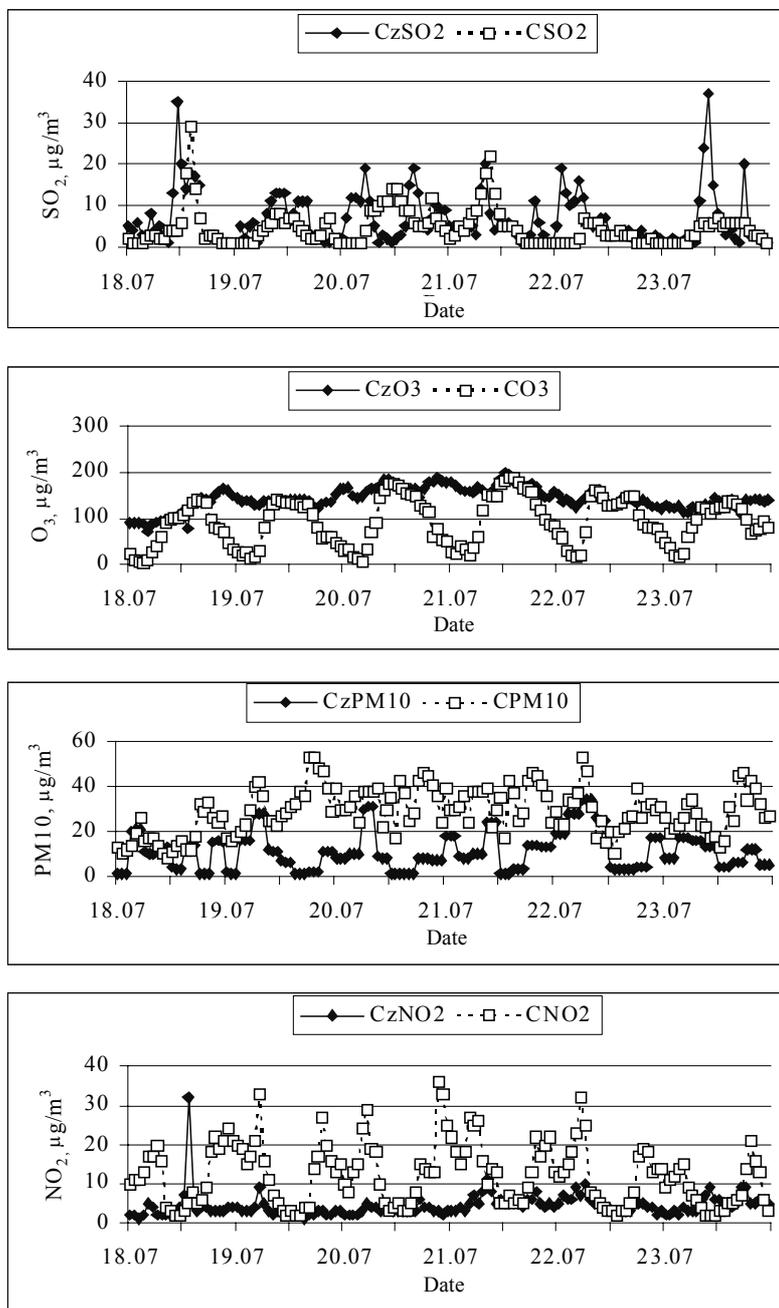


Fig. 5. Time series for basic pollutants monitored in Czerniawa (Cz) and Cieplice (C) during first field studies (18–23 July, 2006)

pollution does not increase during the wintertime in background sites, despite the higher anthropogenic emissions, because of the compensatory effects due to the dispersion conditions. Czerniawa is often above the mixing layer height (MLH) in winters, therefore even in elevated concentrations, pollutants are usually not transported to the higher layers of atmosphere. On the other hand, the weather conditions in Cieplce promote accumulation of pollutants. Because of the lower ventilation factor (MLH and wind velocity – weak air flow and a shallow mixing layer depth) in winter than in summer, the measured concentrations of all pollutants of interest in Cieplce are higher in the winter season, independently of their sources.

Generally, the pollutant concentrations varied widely during all field campaigns. In figure 5, we compared the time series for basic pollutants monitored in Czerniawa (Cz) and Cieplce (C) during the first campaign(18–23 July 2006).

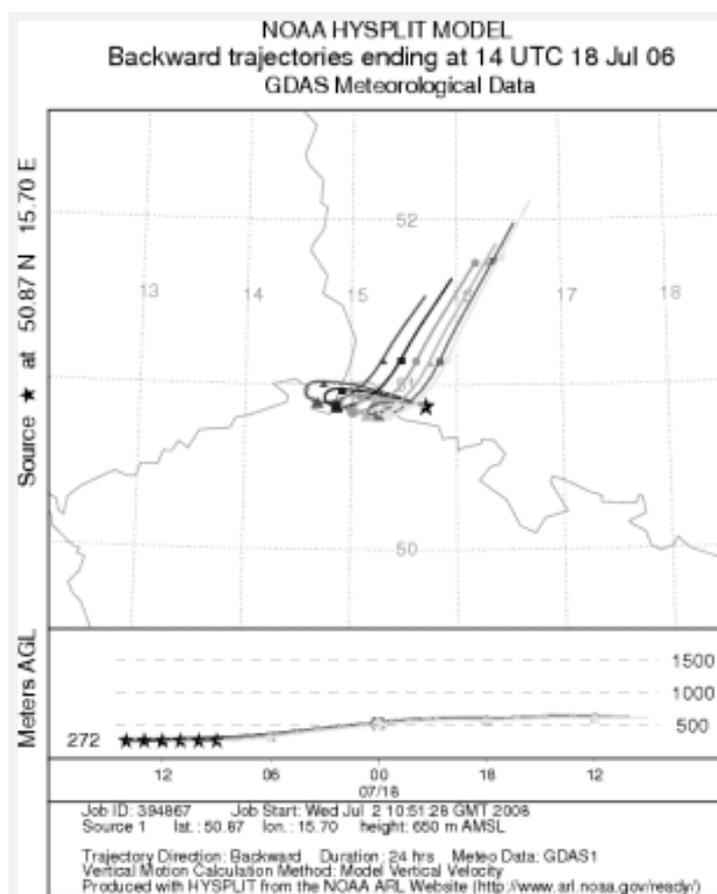


Fig. 6. HYSPLIT 24-h backward trajectories at Czerniawa calculated for hours 09:00–14:00 CET on July 18, 2006

When the air quality is affected by long-range or regional transport, the levels of O<sub>3</sub>, PM10 or other pollutants are coincident with each other and undergo less diurnal variations than in other days. It was confirmed by O<sub>3</sub> and NO<sub>2</sub> behaviour in Czerniawa. However, during specific days “short-lived spikes” of very high SO<sub>2</sub> and NO<sub>2</sub> concentrations were observed in Czerniawa, e.g. on 18 and 23 July. We can recognize the influence of the main stationary source on the pollutant emission, probably smelting operations on July 23, and also on July 18, which manifests itself as the high concentration of SO<sub>2</sub>. High SO<sub>2</sub> concentration was observed just before the noon on 18 July, and the peak in NO<sub>2</sub> and the decrease in O<sub>3</sub> two hours later. The simultaneous decrease in O<sub>3</sub> level at that time testified to the advection of fresh plumes produced by electric power plants. The reaction of background O<sub>3</sub> with generated NO leads to a decrease in O<sub>3</sub> concentration in the “core” of the plume, with concurrent production of NO<sub>2</sub> and the anti-correlation observed. SO<sub>2</sub> concentration was also higher at that time, 14 µg/m<sup>3</sup>, but masked by the previous peak around noon. The overlapping impact of these two emission sources was confirmed by the analysis of trajectories (figure 6). Air masses changed their direction from north-east (50 km – copper smelter) to south-west (20 km – power plant) just around noon. Regional influence in Czerniawa is also evidenced by the data presented in figure 7 which shows the mean 24-hour concentrations of heavy metals at the monitoring stations for 18–23 July 2006. The elevated concentration of Cu and Zn in Czerniawa should be noted. These metals appear episodically and can be assumed as tracers of smelting operations. However, Cu and Zn concentrations are much lower than those reported by LONATI et al. [6] for Milan, 249 and 276.5 ng/m<sup>3</sup>, but broadly consistent with the concentrations in urban areas of the United Kingdom [7].

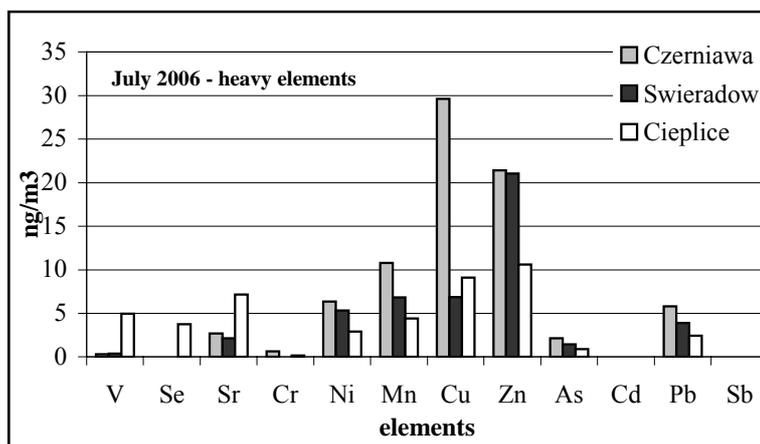


Fig. 7. Mean 24-hour concentrations of heavy metals at monitoring stations for July 18–23, 2006

Local sources are dominant in the case of NO<sub>2</sub> in Cieplice – NO<sub>2</sub> concentrations exhibited typical diurnal variations. This confirms also O<sub>3</sub> patterns – the nocturnal decrease in O<sub>3</sub> concentrations observed in the urban areas was caused by its efficient chemical consumption and deposition and lack of photochemical production of O<sub>3</sub> as well.

#### 4. CONCLUSIONS

Air pollution in the Polish part of the Black Triangle has nowadays diminished significantly compared to the 90's of the last century. Nevertheless, its pollution with PM<sub>10</sub> and heavy metals is believed to be still an actual problem.

Our analysis led to two major conclusions. First, local influence (local pollution sources, fumigation effect) is still evident in Cieplice. Second, Czerniawa generally reflects the regional background pollutant concentrations, but it is also periodically subjected to polluted air coming from regional sources, which contribute to a significant increase in SO<sub>2</sub> and heavy metal levels. During both summer and winter months, PM<sub>10</sub> concentration in Cieplice is higher than that in Czerniawa, particularly in winters. Contrary to Cieplice, PM<sub>10</sub> concentrations in Czerniawa did not show any significant seasonal differences.

#### ACKNOWLEDGEMENT

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TENDENCJE W ZMIANACH STĘŻEŃ ZANIECZYSZCZEŃ  
POWIETRZA ATMOSFERYCZNEGO NA OBSZARZE CZARNEGO TRÓJKĄTA

Dokonano analizy stężeń dwutlenku siarki i pyłu w dwóch wybranych miejscowościach uzdrowiskowych położonych na obszarze styku Czarnego Trójkąta. Przeanalizowano roczne serie i zmienność pyłu, które zmierzono w miesiącach letnich (lipiec) i zimowych (luty) w latach 1996–2007. Dodatkowo, w lipcu 2006, zmierzono stężenia metali ciężkich w pyłe (analiza EDXRF). Potwierdzono, że stężenia zanieczyszczeń powietrza znacznie zmalały w porównaniu z tymi z lat 90-tych ubiegłego wieku.