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*Original Research*

# Statistical Modelling of Changes in Concentrations of Atmospheric $NO_2$ and $SO_2$

**J. Szyda, H. Wierzbicki, A. Stokłosa**

Department of Animal Genetics, Wrocław University of Environmental and Life Sciences,
Kożuchowska 7, 51-631 Wrocław, Poland

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## Abstract

Our study aims at statistical modelling of changes in the level of air pollution in the area of Szczawno Zdrój in Poland throughout the period of 1989-2003, which was marked by a decrease in coal production as well as by rapid increases in traffic intensity. Sulphur dioxide and the nitrogen dioxide were chosen as pollution indicators. Changes of the averaged concentrations across years are modelled by 5 regression models: linear, logarithmic, 2nd degree polynomial, 3rd degree polynomial and 4th degree polynomial. Changes in average yearly concentrations of pollutants during the 1980s and 1990s indicate considerable improvement of air quality regarding sulphur dioxide and nitrogen dioxide contents. A noticeable tendency toward a decrease in air pollution results from limiting of sulphur dioxide emission sources, which translates into liquidation of especially burdensome industrial plants, installation of devices for sulphur removal from fumes, and substantial reduction of air pollution coming from the Czech Republic and Germany.

**Keywords:** air pollution, nitrogen dioxide, regression models, sulphur dioxide

## Introduction

Air pollution is seen by researchers as a serious threat to human health and the environment, though the extent of the problem clearly varies by region. Polluted air contains substances that are hazardous to health. Among the most often observed deficiencies are respiratory problems, lung cancer, chronic obstructive pulmonary disease, weakening of the immune system, and lung function problems [1-3].

A considerable amount of the air pollution in Poland results from $SO_2$, $NO_x$ and $CO_2$ originating from extensive energy use, together with the lack of effective pollution control strategies [4]. About 86% of enterprises harmful for the environment have no installations for gas removal. The high quantity of coal oxides and sulphur oxides is due to burning large quantities of coal.

Coal has long been the basic fuel in the Polish electricity sector. Not only industrial power stations and heat and power generating plants have been responsible for air pollution, but also local boiler rooms and individual stoves have used coal for heat energy production. Solid fuels are difficult to burn in simple combustion devices such as household cooking and heating stoves without substantial emissions of pollutants, principally because of the difficulty of completely premixing the fuel and air during burning, which is done easily with liquid and gaseous fuels [5]. The amount of sulphur in fumes depends on its concentration in burning coal. In Polish coals the sulphur content ranges from 0.32% to 2.28% with the average content of 1.2%. Therefore, the construction of desulphurization installations is necessary. In households that use sulphur-rich coals, sulphur dioxide pollution affects not only indoor air quality but also outdoor air quality at a local or regional scale. Because coal burns at a substantially higher temperature than biomass, higher emissions of $NO_2$ were measured for coal combustion than for biomass combustion [3]. Nitrogen dioxide is an important air pollutant because it contributes to the formation of photochemical smog, which can have a significant impact on human health [6, 7]. The major source of nitrogen dioxide is the burning of fossil fuels: coal, oil and gas. Most of the nitrogen dioxide in cities comes from motor vehicle exhaust (about 50%, 12). Other sources of nitrogen dioxide are petrol and metal refining, electricity generation from coal-fired power stations, other manufacturing industries and food processing. In Poland, the bulk of carbon dioxide emissions has its source in industrial sectors assignable in the IPCC approach to the categories 'energy' and 'industrial processes', whereas the shares of the remaining categories appear negligible [8]. Producing 1 GJ energy from hard coal provokes generation of 94.60 kg of $CO_2$ into the atmosphere [9]. Coal burning also causes generation of coal oxides.

Emissions can be reduced by avoiding the formation of pollutants or by flue gas cleaning. In addition, advanced technologies which lead to an increase of the energy conversion efficiency will generally reduce the specific emissions of all pollutants [10].

In air pollution problems, the air quality models are used to predict concentrations of pollutants in the atmosphere. They form one of the most important components of an urban air quality management plan. Modelling provides the ability to assess current and future air quality in order to enable "informed" policy decisions. Thus, air quality models play an important role in providing information for better and more efficient air quality management planning [11, 12]. Several mathematical models are used in all aspects of urban air quality assessment and prediction [13-17]. These models can be classified into local or mesoscale models. Computational Fluid Dynamics models are generally used for a simulation of local air pollution, whereas mesoscale modelling approaches (simple models, k-based models, Full Reynolds stresses models) and other larger scale models use prognostic equations for the calculation of wind, temperature, moisture content, etc. [1, 19-21].

The Energy Flow Optimization Model – Environment has been widely used to assess the feasibility of various emission reduction targets for $SO_2$ and $NO_x$ and the corresponding costs in Western European countries [22] as well as in Central and Eastern European countries [23]. In recent years new forecast models (LDF - Fisher Linear Discriminant Function, QDF - Quadratic Discriminant Function, REGF - Regression Function, BPNN - Backprop Neural Network, and RBFN - Radial Basis Function Network) for tacking air quality have been developed [1]. The prediction of the pollution levels can also be carried out using the stochastic method known as kriging. Stochastic methods presuppose that different measurements of the pollutant in the area under study, that is, the samples taken, actually correspond to the distribution of a single random variable distributed spatially in that area [6].

In our study the region of Szczawno Zdrój was chosen as a study area for modelling the concentrations of air pollution. The town is located in the region of coal mining industry, power generation and district heating plants and considerable traffic. Moreover, the analysis period of 1989-2003 was marked by a decrease in coal production as well as by rapid increase in traffic intensity. Szczawno Zdrój is strongly affected by the industrial environment of the nearby town of Wałbrzych. For many years this region has been recognized as one of the most industrialized regions in Poland. In the 1980s Szczawno Zdrój was an important centre of coal mining with three large mines. Because of their nonprofitability, since the mid-1990s the mines had been successively shut down so that in August 1998 the last of the coal mine was closed.

The present study aims at statistical modelling of changes in the concentration of air pollution in Szczawno Zdrój, based on data published by the Regional Department of Environmental Protection [24]. Changes of the averaged concentrations of air pollutants across years were modelled by fitting different regression models. The sulphur dioxide ($SO_2$) and the nitrogen dioxide ($NO_2$) are chosen as pollution indicators, because:

(i) the evidence that $SO_2$ and $NO_2$ are positively and significantly associated with mortality [25],

(ii) the atmospheric concentrations of $SO_2$ and $NO_2$ have constantly been measured and recorded in the data base over the analyzed period,

(iii) the data are freely available from the Regional Department of Environment Protection.

The study considers a time span between 1989 and 2003, which covers the period of the restructurization of coal mines and enhanced industrial growth of the investigated region, thus enabling visualization of the effect of economic transformation and change in environmental policy on air quality. The paper is divided into five parts. Following the introduction, the next section describes the region under investigation and statistical modelling of the data. Further results are reported and discussed in the following section. The last section summarizes the study.

## Experimental Procedures

### Study Area

The town of Szczawno Zdrój, situated in southwestern Poland (Lower Silesia), is characterized by a well developed road network and shelters a number of large and small-scale industries emitting many pollutants such as CO, $CO_2$, $SO_2$, PM10 and $NO_x$ (Fig. 1). The geographical position of the town (50° 47' 58" N; 16° 15' 18" E) as well as its climate conditions cause frequent fog and persistent fog. Szczawno Zdrój is located 410-420 meters above sea level, in the very heart of the Wałbrzyskie Mountains, surrounded by the slopes of Gedymina Hill (height: 517 meters) and Chełmiec Hill (height: 869 meters). This results in poor air circulation. The town features specific microclimate with a frequently formed inversion layer. Additionally, the town area (15 $km^2$) is characterized by compactness of residential housing, inappropriate setting of industrial plants and intensive traffic. All these factors are responsible for the accumulation of pollutants in the atmosphere.

Propagation of pollution across the region of Szczawno Zdrój is strongly influenced by local geographical conditions, i.e. location of particular cities and towns in small valleys surrounded by natural hills and dumps, which originate from coal mining. Such a landscape hinders the natural process of clearing the atmosphere of dusts and gases through gravity-induced air circulation. Compact architecture within the city and relatively weak winds further impede natural air circulation. Moreover, the propagation of toxic gases and dust is accelerated by the location of major factories in the direction of the most common winds. Recently, a significant decrease in industrial emissions has been observed, but the still high level of air pollution is attributed to the so-called “low emissions” from the municipal sector, i.e. outdated boiler houses and households still using coal and coke for heating, which is especially profound during the heating season (i.e. autumn and winter). Szczawno Zdrój is located along a highway and thus the town is also subjected to strong influence of air pollution resulting from traffic. The whole region of Lower Saxony, especially its southwestern part, is additionally affected by air pollution from Germany and the Czech Republic.

## Statistical Analysis

In the analysis, averaged year concentrations of $SO_2$ and $NO_2$, calculated as arithmetic daily averages (24 h) concentrations measured by two stationary stations available for monitoring air pollution within Szczawno Zdrój area, are considered. One of the stations (station 1) is located close to the town centre and the other one (station 2) is in the residential area. Both stations assess daily concentrations of $SO_2$ using the method following the national standard (PN-76/2-04104/01) and $NO_2$ by modified Saltzman approach [26].

Changes of the averaged concentrations across years are described by the following regression models, whose parameters ($\mu$, $\beta_i$) are estimated using the least squares procedure:

a) linear: $y = \mu + \beta_1 x$,
b) logarithmic: $y = \mu + \beta_1 \ln(x)$,
c) 2nd degree polynomial: $y = \mu + \beta_1 x + \beta_2 x^2$,
d) 3rd degree polynomial: $y = \mu + \beta_1 x + \beta_2 x^2 + \beta_3 x^3$,
e) 4th degree polynomial: $y = \mu + \beta_1 x + \beta_2 x^2 + \beta_3 x^3 + \beta_4 x^4$,

...where $y$ represents concentration ($SO_2$ or $NO_2$), $x$ - year (from 1989 to 2003), $\mu$ - the intercept, and $\beta_j$ is the regression coefficient corresponding to the $j$-th degree. The function best describing concentration changes with the smallest number of coefficients is chosen based on the mean square error criterion (MSE):

$$MSE = \frac{\sum_{i=1}^{N} (y_i - \hat{y}_i)^2}{N - p}$$

...where: $y_i$ and $\hat{y}_i$ are respectively an observed concentration and a concentration predicted by the model (a-e) for the $i$-th

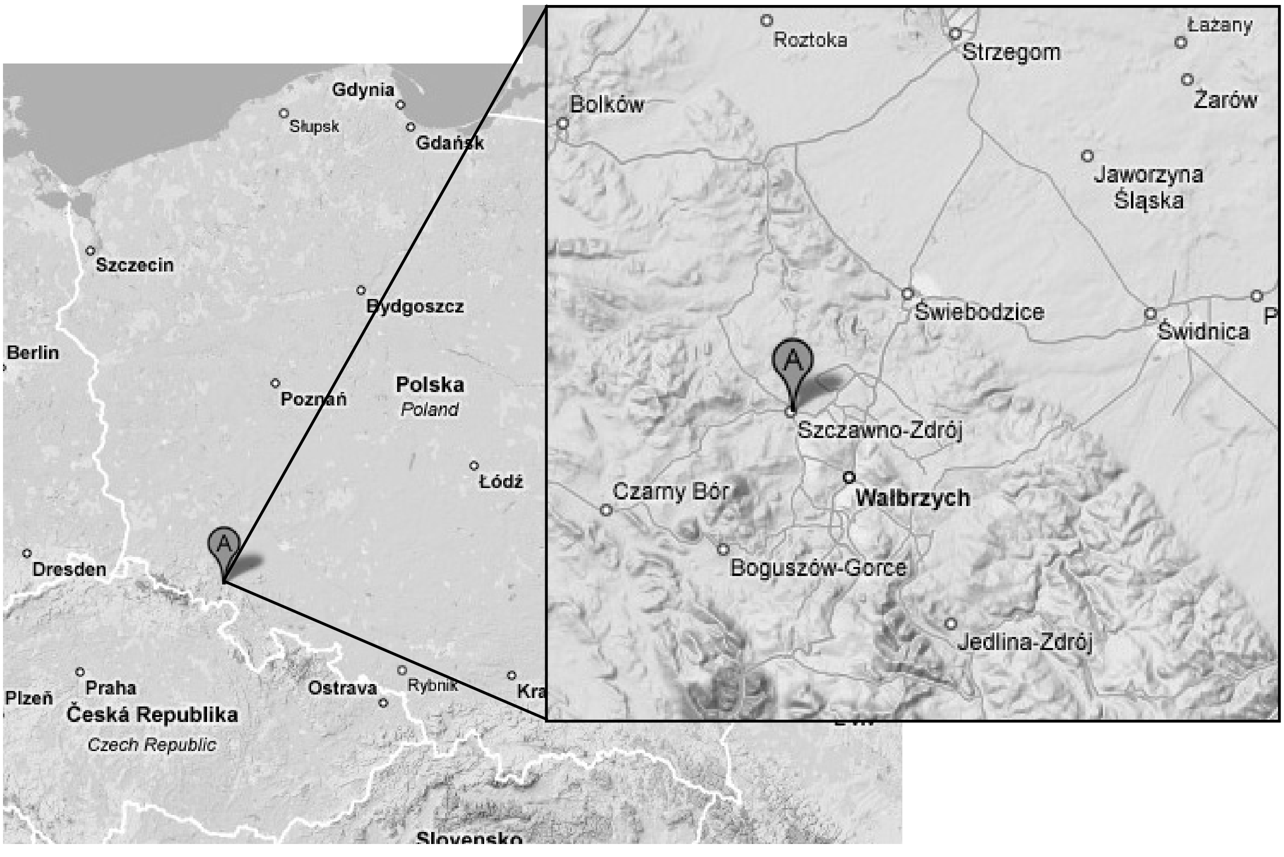

Fig. 1. The geographical localization of Szczawno Zdrój.

year, $N$ is the total number of observations (i.e. years with measurements available) and $p$ is the number of parameters in the model (i.e. $\mu$ and $\beta$) [27].

Additionally, the differences in measurement variances between station 1 and station 2 are tested using:

$$F = \frac{\sigma_j^2}{\sigma_{j'}^2} \sim F_{N-1,N-1}$$

...with corresponding hypotheses expressed by $H_0{:}\sigma_j^2 = \sigma_{j'}^2$ and $H_1{:}\sigma_j^2 \neq \sigma_{j'}^2$, where $\sigma_j^2$ and $\sigma_{j'}^2$ denoting respectively a higher and a lower variance of measurements.

## Results and Discussion

Regression curves fitted to the $SO_2$ and $NO_2$ data are shown in Figs. 2-5. The models characterized by the lowest MSE value are represented by a trick solid curve.

The highest concentrations of $SO_2$ had been recorded from 1989 to 1993, i.e. when coal mines, coking plants and most other industrial plants were active. Based on the MSE criterion the best models describing $SO_2$ concentrations are the logarithmic and 2nd degree polynomial for stations 1 and 2, respectively (Table 1). More features of pollution concentration changes across years can be revealed by detailed interpretation of parameters of the fitted regression models. Following the simplest assumption of a linear decrease in concentration, it is possible to notice that the decrease in concentration of $SO_2$ amounts to 2.9 $\mu g/m^3$ each year as measured by station 1 and to 4.9 $\mu g/m^3$ by station 2. The same pattern is recorded assuming a continuous, but nonlinear, decrease described by the logarithmic curve, where the estimated parameters measure the absolute change in concentration given the relative change in time. Here, 1% increase in time relates to the estimated decrease in $SO_2$ concentration by 0.2 $\mu g/m^3$ (station 1) and by 0.3 $\mu g/m^3$ (station 2). Still, for $SO_2$ concentration measured by station located

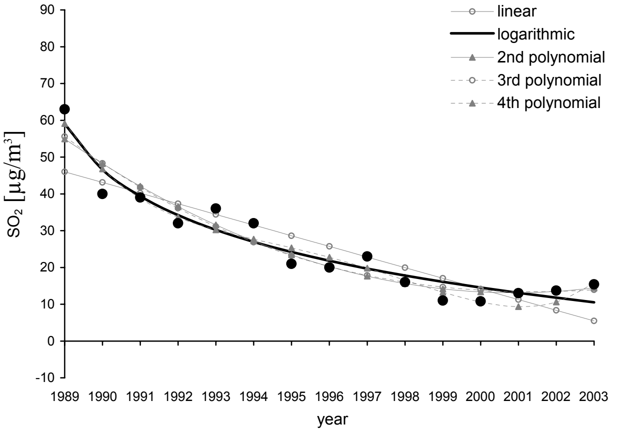

Fig. 2. Observed (●) and fitted (lines) yearly concentration of $SO_2$ as measured by station 1. The best fitting line is marked by a thick solid line (**—**).

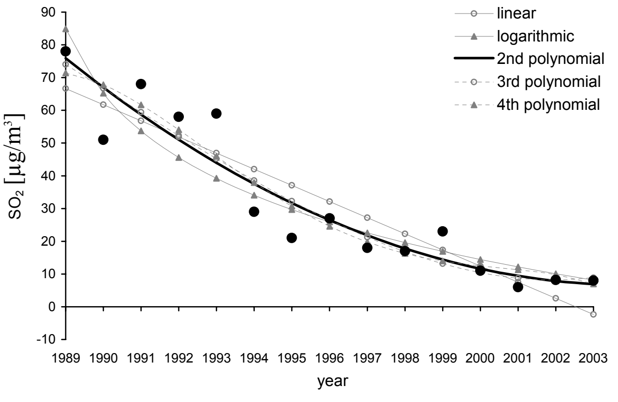

Fig. 3. Observed (●) and fitted (lines) yearly concentration of $SO_2$ as measured by station 2. The best fitting line is marked by a thick solid line (**—**).

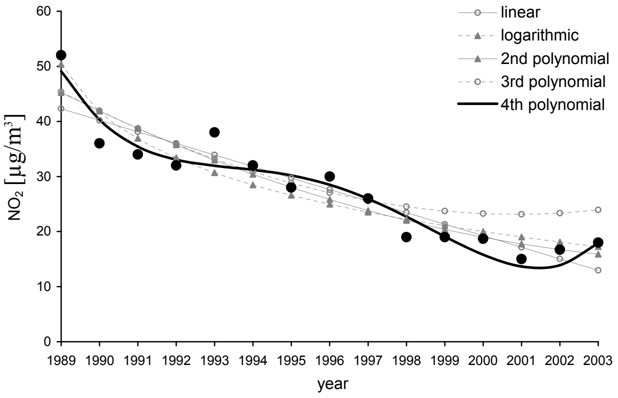

Fig. 4. Observed (●) and fitted (lines) yearly concentration of $NO_2$ as measured by station 1. The best fitting line is marked by a thick solid line (**—**).

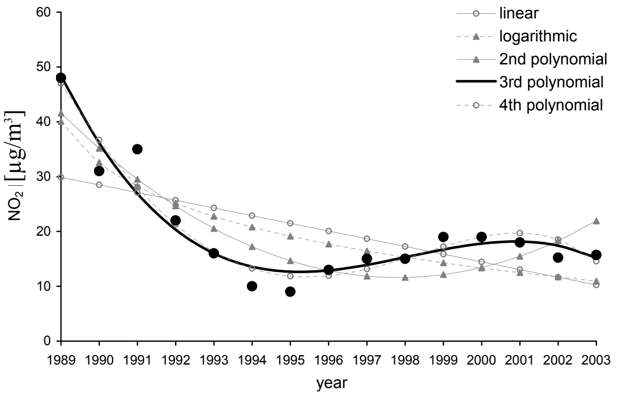

Fig. 5. Observed (●) and fitted (lines) yearly concentration of $NO_2$ as measured by station 2. The best fitting line is marked by a thick solid line (**—**).

Table 1. Summary of the parameters and quality (measured by MSE) of the fitted models. Parameters corresponding to the best fitting model are marked in bold.

| Fitted model | Station 1 | | Station 2 | |
|---|---|---|---|---|
| | MSE | Parameter estimates | MSE | Parameter estimates |
| | $SO_2$ | | | |
| linear | 47.4 | $\hat{\mu} = 48.9, \hat{\beta}_1 = -2.9$ | 99.6 | $\hat{\mu} = 71.6, \hat{\beta}_1 = -4.9$ |
| logarithmic | **16.8** | $\hat{\mu} = \mathbf{59.0}, \hat{\beta}_1 = \mathbf{-17.9}$ | 93.8 | $\hat{\mu} = 84.9, \hat{\beta}_1 = -28.3$ |
| 2nd polynomial | 21.5 | $\hat{\mu} = 62.2, \hat{\beta}_1 = -7.6, \hat{\beta}_2 = 0.3$ | **5.9** | $\hat{\mu} = \mathbf{85.4}, \hat{\beta}_1 = \mathbf{-9.8}, \hat{\beta}_2 = \mathbf{0.3}$ |
| 3rd polynomial | 23.3 | $\hat{\mu} = 63.7, \hat{\beta}_1 = -8.5,$<br>$\hat{\beta}_2 = 0.4, \hat{\beta}_3 = -0.006$ | 81.2 | $\hat{\mu} = 81.1, \hat{\beta}_1 = -7.0,$<br>$\hat{\beta}_2 = -0.11, \hat{\beta}_3 = 0.02$ |
| 4th polynomial | 18.1 | $\hat{\mu} = 77.4, \hat{\beta}_1 = -21.9,$<br>$\hat{\beta}_2 = 3.9, \hat{\beta}_3 = -0.3, \hat{\beta}_4 = 0.01$ | 85.2 | $\hat{\mu} = 71.3, \hat{\beta}_1 = 2.5,$<br>$\hat{\beta}_2 = -2.6, \hat{\beta}_3 = 0.3, \hat{\beta}_4 = -0.007$ |
| | $NO_2$ | | | |
| linear | 17.6 | $\hat{\mu} = 44.4, \hat{\beta}_1 = -2.1$ | 74.3 | $\hat{\mu} = 31.2, \hat{\beta}_1 = -1.4$ |
| logarithmic | 14.0 | $\hat{\mu} = 50.3, \hat{\beta}_1 = -12.2$ | 40.8 | $\hat{\mu} = 40.0, \hat{\beta}_1 = -10.7$ |
| 2nd polynomial | 15.8 | $\hat{\mu} = 48.8, \hat{\beta}_1 = -3.6, \hat{\beta}_2 = 0.1$ | 29.6 | $\hat{\mu} = 48.7, \hat{\beta}_1 = -7.6, \hat{\beta}_2 = 0.4$ |
| 3rd polynomial | 33.5 | $\hat{\mu} = 49.1, \hat{\beta}_1 = -3.8,$<br>$\hat{\beta}_2 = 0.1, \hat{\beta}_3 = 0.001$ | **12.2** | $\hat{\mu} = \mathbf{63.9}, \hat{\beta}_1 = \mathbf{-17.4},$<br>$\hat{\beta}_2 = \mathbf{1.9}, \hat{\beta}_3 = \mathbf{-0.1}$ |
| 4th polynomial | **10.6** | $\hat{\mu} = \mathbf{63.5}, \hat{\beta}_1 = \mathbf{-17.8},$<br>$\hat{\beta}_2 = \mathbf{3.8}, \hat{\beta}_3 = \mathbf{-0.3}, \hat{\beta}_4 = \mathbf{0.01}$ | 12.4 | $\hat{\mu} = 58.7, \hat{\beta}_1 = -12.4,$<br>$\hat{\beta}_2 = 0.6, \hat{\beta}_3 = 0.1, \hat{\beta}_4 = -0.004$ |

within the residential area (station 2), changes in pollution regarding the studied period are too complex to be sufficiently described by linear or logarithmic functions and thus require models with more parameters. A 2nd degree polynomial applied to $SO_2$ data from station 2 shows that minimum pollution is predicted for 2004, meaning that throughout the periods of 1989-2003 there has been a nonlinear decrease in $SO_2$ concentration with a faster decrease at the beginning than at the end of the measurement period. The reduction in $SO_2$ concentration within this period coincides with the fact that coal mines have subsequently been closed, while big industrial plants have either improved their methods of purifying fumes or have also been closed. Moreover, at that time the air pollution from the The Czech Republic and Germany has gradually decreased [28]. A similar decreasing trend has also been observed in other European cities [29]. The comparison of the variances of concentrations recorded by each station, with the use of the F test, reveals significant differences between the stations in recorded $SO_2$ concentration changes.

Assuming the simplest, linear change in $NO_2$ concentration reveals evidence of decrease by 2.1 µg/m$^3$ (station 1) and 1.4 µg/m$^3$ (station 2), which is slower than observed for $SO_2$. However, changes in $NO_2$ concentration have been more complex, so that for measurements at station 1, at least four parameters are required to fit the data – indicating three major changes in $NO_2$ concentration. In particular, we observe a relatively rapid decrease between 1989 and 1993, then a slightly slower decrease between 1993 and 1998, then again a rapid decrease after 1998, until 2001, when the minimum concentration is observed, followed by an increase in concentration in the last two years examined in the study (2002 and 2003). $NO_2$ values obtained by station 2 also show a similar tendency of a fast decrease in concentration only until 1995, but afterwards, differently than concentrations recorded by station 1, an increase till 2001 is observed, followed by decreasing concentration at the very end of the measurement period. A significant decrease in $NO_2$ concentration can be observed in the first part of the studied period. A continuous increase in the number of vehicles has resulted however, in an increase of $NO_2$ emission in the second part of the studied period. A similar, close relationship between traffic intensity and $NO_2$ concentration has similarly been reported for other cities, e.g. for Copenhagen [30]. No differences in overall $NO_2$ concentration variability between both stations were observed.

## Conclusion

Generally, adecreasing tendency in emissions can be recognized in all graphs. $SO_2$ measurements show less variation than $NO_2$ measurements, since the former can be described by simpler curves (i.e. curves with fewer parameters).

Although only a particular location was studied in this paper, it was chosen as a model town representative for the whole area. Thus, we believe our conclusions can be extended to the whole region of Wałbrzych, which had been subjected to investment in coal mines and power plants in the past, which nowadays are being replaced by alternative sources of energy and modern industry. In general, the replacement process leads to the improvement of air quality. The lower emission of $SO_2$ has been observed throughout the whole studied period as well as predicted for future years by the course of regression curves. However, the recent concentration of $NO_2$ shows an increasing tendency that probably results from a higher number of vehicles and increased traffic within the studied region.

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