The methods of forecasting of SO$_2$ and suspended dust concentrations for warning purposes in the example of selected polluted regions in Poland

D. Foszcz, T. Niedoba & J. Siewior

$^1$Department of Mining and Geoengineering, University of Science and Technology in Krakow, Poland
$^2$Provincial Inspectorate of Environment Protection, Katowice, Poland

Abstract

The problems of air pollution monitoring is presented in this paper because of its harmful influence on the natural environment and the health conditions of people. In the example of the city of Krakow and cities of the Upper Silesia Industrial Area, the state of air pollution by suspended dust and SO$_2$ were characterised. Statistical analysis of the data from automatic monitoring stations from the region mentioned above was performed and analysis of dependencies between the state of air pollution and meteorological data was carried out. The methodology of pollution modelling for the warning purposes was discussed.

In this paper, on the basis of the measurement data from automatic monitoring stations, the forecast models of SO$_2$ concentration and suspended dust were determined (ARX models and models obtained from adaptive modelling), which can be generally used for the purposes mentioned above.

Keywords: air pollution monitoring, statistical methods, sulphur dioxide, suspended dust, ARX models, adaptive models.

1 Introduction

Sulphur dioxide (SO$_2$) is a colourless, reactive gas that is odourless at low concentrations but has a pungent smell at very high concentrations. Sulphur dioxide is highly soluble in water, resulting in the formation of sulphurous acid (VI). This can be done by homogenous oxidation and reactions in solution, with
or without catalytic factor. It reacts on the surface of a variety of airborne solid particles and can be oxidised within airborne water droplets.

Sulphur dioxide, SO$_2$, enters the atmosphere as a result of both natural phenomena and anthropogenic activities e.g.: combustion of fossil fuels; oxidation of organic material in soils; volcanic eruptions; biomass burning.

Effects of volcanic eruptions may have an impact on air traffic, as such eruptions are important sources of ash (aerosols) and sulphur dioxide in the atmosphere. However, emissions are sporadic, intermittent and often occur in uninhabited regions.

Mainly power stations, combustion of fuel and vehicles are the anthropogenic sources of SO$_2$ pollution. Coal burning is the single largest man-made source of sulphur dioxide, accounting for about 50% of annual global emissions (e.g. from 1 tone of hard/brown carbon of 3% of sulphur 60 kg of SO$_2$ is leaking), with oil burning accounting for a further 25 to 30%.

Changes in the abundance of sulphur dioxide have an impact on atmospheric chemistry and on the radiation field, and hence on the climate. Consequently, global observations of sulphur dioxide are important for atmospheric and climate research.

The major health concerns associated with exposure to high concentrations of sulphur dioxide include effects on breathing, respiratory illness, alterations in pulmonary defenses, and aggravation of existing cardiovascular disease. In the atmosphere, sulphur dioxide mixes with water vapour producing sulphuric acid. This acidic pollution can be transported by wind over many hundreds of miles, and deposited as acid rain (www.temis.nl, [1], Szczepaniec-Cieciak and Koscielniak [2]).

According to the paper (Rutowski et al, [3]) it is proved that exposure of human beings to SO$_2$ results in increasing of total number of lymphocytes (the tests were carried on by using Amaya-Sugiura passive sampling and ion spectrophotometry)

The harmful influence of SO$_2$ depends on both of concentration and time of exposure, what is presented on fig. 1.

It occurs that even a small concentration of SO$_2$ in situation of longer exposure to people causes disadvantageous effect on health, mainly connected with respiratory and blood systems (Rutowski et al, [3]).

Aerosols are tiny particles suspended in the air. Some occur naturally, originating from volcanoes, dust storms, forest and grassland fires, living vegetation, and sea spray. Human activities, such as the burning of fossil fuels, prescribed fires, and the alteration of natural surface cover, also generate aerosols. Many human-produced aerosols are small enough to be inhaled, so they present a serious health hazard around industrial centers or even hundreds of miles downwind. Another unfortunate aspect of the small sizes of respirable dusts is that they have a very low settling velocity and can remain suspended in air indefinitely. Additionally, thick dust or smoke plumes severely limit visibility and can make it hazardous to travel by air or road. New research shows that aerosol pollution can modify cloud properties to reduce or prevent precipitation.
in the polluted region. Aerosol containing black carbon can impact climate and possibly reduce formation of clouds (Carver, [4]).

Sources of atmospheric dust are winds blowing over dry earth (plowed fields, deserts, and roads), the various products of combustion, volcanic eruptions, salt spray from the oceans, pollen and other material from plants, and meteoric particles. The detonation of nuclear devices in the atmosphere creates radioactive dust.

Dust sometimes settles quickly on surfaces, but vast quantities are carried to the upper layers of the air and suspended there for long periods of time.

Classification of dusts may be divided into five categories:

- **Toxic dusts** – these can cause chemical reactions within the respiratory system or allow toxic compounds to be absorbed into the bloodstream through the alveolar walls. They are poisonous to body tissue or to specific organs (Walli, [5]).
- **Carcinogenic (cancer causing) dusts**;
- **Fibrogenic dusts**;
- **Explosive dusts**;
- **Nuisance dusts**.

The Silesian province, as well the region of Krakow are ones of the most polluted regions in Poland. So, the forecasting and researches over SO₂ and dusts concentrations in this area are crucial and very important.

### 2 Methodology of pollution spreading modelling

Modelling of air pollution spreading bases on a reciprocal connection between the features conditioning dispersion of pollution. Among these features,
influencing on expansion of pollutants, as: meteorological conditions, source and emission characteristics, reciprocal affection between pollution and ground surface, the most important are meteorological conditions. So, the modelling of atmospheric phenomenons is crucial. Generally, dispersive models may be divided into the categories presented on fig. 2. (Chodorski and Pietrzyk, [6]).

**Figure 2: Division of dispersion models.**

Diffusive models are based on physical laws of pollution propagation by hovering and diffusion, derived with various simplifying assumptions. In these models both differential equations and their coefficients of certain physical interpretation were applied [Chodorski and Pietrzyk, [6]; Borysiewicz and Stankiewicz, [7]; Juda and Chrosciel, [8]).

Gauss models are based on the assumption that the concentrations distribution function is a normal distribution with variances depended on distance from a source and atmosphere stratification. The models of “k theory” assume that the turbulent concentrations flow is proportional to mean concentration. This assumption is called the closing of first extent.

The point of start to the theoretical researches of pollution spreading in atmosphere are differential equations of diffusion in moving centre. The solution of these equations allow to determine the pollution concentration in any distance from a source and (for the point of pollution emission – a chimney) its height assuring optimal spreading of emitted pollution. The basic assumption in these equations is that the spreading is symmetrical round the „z” axis.

The general form of equation of turbulent diffusion in moving centre, being applied in further considerations, in rectangular co-ordinate system is presented by the following equation:

\[
\frac{dS}{d\tau} = \text{div}(KVS) - S\text{div}\bar{c} + f
\]  

where: \(S\) – gas or dust pollution concentration, \(K\) – coefficient of atmospheric diffusion, \(\bar{c}\) – vector of air movement velocity, \(f\) – a function expressing the action of volumetric emission source of pollution (decrease or increase of pollution).

Depending on the assumptions and simplifications we obtain various forms of solution of diffusion equation. As the most known and rated to the basic ones the solutions given by Sutton, Pasquille, Bosanquet, Pearson and Calder are considered, which differ among themselves by assumed vertical and horizontal diffusion coefficients. In the equations mentioned above the independence of atmosphere turbulence coefficients on co-ordinates \(x, y, z\) is assumed. The given
solutions of diffusion equation are the base for the analytic solutions (Juda and Chrosciel, [8]).

The starting point to solving the diffusion equation in moving centre by Pasquille was the introduction of atmospheric diffusion coefficients, given by equations:

\[ \sigma_x = \sqrt{\bar{u}^2} \frac{x}{\bar{u}} = T_x x, \quad \sigma_y = \sqrt{\bar{v}^2} \frac{x}{\bar{u}} = T_y x, \quad \sigma_z = \sqrt{\bar{w}^2} \frac{x}{\bar{u}} = T_z x \]  

(2)

where: \( \sigma_x, \sigma_y, \sigma_z \) – atmospheric diffusion coefficients of Pasquille [m], \( \bar{u}^2, \bar{v}^2, \bar{w}^2 \) – variances of wind velocity in direction x, y, z resulting from the existence of turbulent movements [m²/s], \( \bar{u} \) – mean wind velocity in considered layer [m/s], \( T_x, T_y, T_z \) – measures of intensivity of turbulence in directions x, y, z (dimensionless quantities).

The diffusion coefficients given by Pasquille are depend on the distance x from the source of emission, it means on the time of pollution dislocation, which is expressed by the ratio \( x/\bar{u} \). The numerical values of coefficients in directions y and z depend generally on atmosphere’s turbulence structure, height above the surface of earth, roughness of terrain and its topography, time of concentration, distance from the source of emission and the state of atmosphere’s equilibrium. Pasquille gave the key to determine the state of atmosphere’s equilibrium, allowing the determination of coefficients \( \sigma_y \) i \( \sigma_z \) for assumed distance from the source of emission.

For the purpose of simplifying the general solution of diffusion equation Pasquille introduced the following assumptions:

1. The field of concentration is determined in time: \( \partial S/\partial \tau = 0 \). To fulfil this condition, the intensity of emission must be constant in time, no shorter than the time of consideration of pollution movement from source to the point, where we determine the pollution concentration. Also, the meteorological conditions should be constant in this time.

2. The components of air movement velocity in direction y and z are equal to zero \( v = w = 0 \),

3. The air is treated as non-compressible fluid,

4. The atmospheric diffusion coefficients do not depend on height above the surface of earth.

5. There are not volumetric sources of emission, what means that \( f = 0 \).

Thanks to these assumptions, after applying the air turbulence \( \sigma_i = \sqrt{2K_i \frac{x}{\bar{u}}} \) for the directions y and z, where \( x/\bar{u} \) is the time of pollution movement \( \tau \), we obtain the differential equation:

\[ \frac{\partial S}{\partial \tau} = \frac{d}{d\tau} \left( \frac{\sigma_y^2}{2} \right) \frac{\partial^2 S}{\partial y^2} + \frac{d}{d\tau} \left( \frac{\sigma_z^2}{2} \right) \frac{\partial^2 S}{\partial z^2} = 0 \]  

(3)
Different from the starting equation we interpret the $\partial S/\partial \tau$, assuming that the point of reference is moving with velocity $\bar{u}$ and during this wander it is changing in time. The solution of equation (3) lead to the group of formulaes of Pasquille’s type, obtaining different form, dependly on assumed conditions, i.e. by: $\tau \to 0$, $\frac{E}{u} \delta(y)\delta(H-z)$ we obtain:

$$S = \frac{E}{2\pi \sigma_x \sigma_y \bar{u}} \exp\left(-\frac{y^2}{2\sigma_y^2}\right) \exp\left[-\frac{(z-H)^2}{2\sigma_z^2}\right] + f(u) \exp\left[-\frac{(z+H)^2}{2\sigma_z^2}\right]$$

where: $S$ – concentration of pollution in point of coordinates $x$, $y$, $z$, $E$ – mass intensity of gas pollution emission, $\bar{u}$ – mean value of wind velocity in propagation layer, $H$ – height of point location of emission source, $f(u)$ – function of „reflection” $f(u) = 1$ for wind above 7 m/s and $f(u) = u/7$ for winds below 7 m/s.

From assumptions taken by Pasquille the limitations of application of his equation for open, flat suburban areas occur, when there is not inversion layer. These limitations caused some sort of modifications allowing enlargement of Pasquille’s equation application (Juda and Chrosciel, [8]).

Additional elements, which should be taken into consideration, applying the solution proposed by Pasquille, are decrease of pollution in atmosphere and changeability in time of emission intensity, occurring for unstable sources of pollution emission.

The point of start to diffusion equation calculation of dust concentration is identical as for gas. The differences, which occur during solving the equation may be as following:

The dust grains have an additional vertical falling velocity what for gas components may be ignored even for the biggest molecules.

There may be an assumption, that the dust grains after touching the surface of earth are completely or partially snatched away from aerosol flow (introduction of suitable edge condition for diffusion differential equation).

Because of the fact that the processes of spreading and decreasing of pollution are connected with diffusion and convection gas phenomenons, which are described by partial differential equations of parabolic type, the concept of solving these equations by variation methods with division of selected areas was presented by Ziemkiewicz (Chodorski and Pietrzyk, [6]). In the base of approach proposed by him, the group of solutions for warmth transport in solids was developed. Also, the works, which purpose is to adapt this solution to gas diffusion and convection problems, are undertaken, as well works over proposal of model in base of finite elements, allowing simulation of pollution spreading round the emission source.

The volumetric models are based on the mass balance, taking into consideration only carriage, without pollution diffusion. The Brungfelt model may be rated to the models of this type. The diffusion factor is not also directly taken into consideration in non-diffusive models, based on the formulaes obtained by mathematical statistics or probability theory. The stochastic models are based on the analysis of random character of pollution spreading. The
regressive models are included to them, as they take into consideration the influence of various meteorological elements on the change of mean pollution coefficients values in certain area. It is very desirable to have the possibility to interpret the form and coefficients of equation by the full physical, consistent with phenomenon’s mechanism interpretation (Morawska-Horawska and Tumidajski, [9]).

The SO2 concentration measurements, apart from applying it to air pollution evaluation, should be applied also in the SO2 management by its delimitation (excluding of some objects) during crisis days and also to preventive purposes. To complete these tasks, the mathematical models of pollution forecasts are needed, which should allow to predict sufficiently precisely the values of concentrations and mainly the situations, when the allowed limits are exceeded. It is not necessary to apply the complicate diffusive models to this purpose (Juda and Chrosciel, [8]). Usually, the regressive models are efficient, which forms may have some theoretical reasons. More than once, the application of diffusive models, because of the necessity of having essential data to solve them (quantity of emission, circumstances of spreading as: air diffusion vertical and horizontal coefficients, topography of terrain), is even impossible. Good solution is application of stochastic models, which often give good prediction results, what makes possible the realisation of warning tasks.

To these models we may include (P(t)):

a) model based on determination of SO2 concentration. During 24h the SO2 concentration is the ratio between the mass of emitted SO2 on certain area, mass of drifted SO2 from neighbouring emitters and mass of SO2, which was earlier in this area to the air volume, in which these masses are spreading. Assuming that (for certain direction of air circulation) the mass of first two components depends on temperature (amount of combusted carbon) and chemical changes, connected with air humidity we may write that:  

\[ m = AW^{a}T^{b} \]

where \( A \) is constant and indexes \( a \) and \( b \) characterise the influence of humidity on change of SO2, dependly on temperature. The volume of air layer, where mixing occurs may be expressed by equation:

\[ V = BH^{c}v^{d}, \]

where \( B \) – constant, which characterise the width of mixing layer (constant for certain directions, \( H \) – height of mixing layer, \( v \) – wind velocity; \( c \), \( d \) – indexes connected with the height or velocity corrections The quantities \( H, T, W \) are the functions of \( t \) variable.

If we are taking into consideration the aspect of autocorrelation of SO2 concentrations, we may introduce the correction \( kSO_{2}(t-1) + p \) into the model, which will correct actual concentration \( P(t) \). So, we have:

\[ \hat{P}(t) = \frac{AW^{a}T^{b}}{BH^{c}v^{d}} + kSO_{2}(t-1) + p \]  \hspace{1cm} (5)

A similar model, which, however, does not take into consideration relative humidity and autocorrelation of SO2 concentrations, was presented by Bringfelt (Bringfelt, [10]).
b) Wien model. To forecast SO\textsubscript{2} concentrations in city centre of Wien, the following model is being applied [Bolzern, [11])\[\]

\[\hat{P}(t) = a_0 P(t-1) + a_2 \left[ T(t) + a_3 \right]^{-\alpha} + a_n \left[ v(t) + a_j \right]^{-\beta} \tag{6}\]

The SO\textsubscript{2} concentration in day of number \(t\) is dependent on SO\textsubscript{2} concentration from the previous day, predicted mean temperature \(T(t)\) and predicted mean wind velocity \(v(t)\). The SO\textsubscript{2} concentration is to some extent connected (concentrations autocorrelation) with mass of pollution existing in the area at the end of \((t-1)\)th day. Second expression takes into consideration the increase of SO\textsubscript{2} emission, originated from the heating system, which is „produced” by falling of temperature, and the third expression describes the decrease of SO\textsubscript{2} concentration by the wind (eventually the drifting of neighboring areas).

c) Regressive models. In the range of air pollution concentration forecasting, usually the ARX models are being applied. These models are taking into consideration concentrations autocorrelation (AR) (X – egzogenic points), not forgetting about the meteorological factors and their displacements in time. So, for SO\textsubscript{2} [Morawska-Horawska and Tumidajski, [9]; Finzi and Tebaldi,[12]; Morawska-Horawska and Kuros, [13]) we have:

\[\hat{P}(t) = a_1 P(t-1) + \ldots + a_p P(t-p) + b_0 T(t) + b_1 T(t-1) + \ldots + b_k T(t-k) + c_0 v(t) + c_1 v(t-1) + \ldots + c_l v(t-l) + d_0 W(t) + d_1 W(t-1) + \ldots + d_r W(t-r) + e \tag{7}\]

(the symbols for variables are the same as in previous models)

Usually \(p\) is smaller or equal to 2 and the values \(k, l, r\) do not exceed 1.

d) Adaptive models. Because it is not always possible to transfer the autoregressive model from certain heating season to the next one, it is necessary to actualise the coefficients in base of actually registered results. This method of modelling is called adaptive modelling and is being applied in neural networks (Rastrigin, [14]).

It is assumed, that the atmospheric pollution forecasting model is in the following form:

\[\hat{y} = f(X.C) = \sum_{j=1}^{k} c_j \varphi_j(X) \tag{8}\]

where: \(X\) - vector of independent values, \(C\) – vector of coefficients, \(j = 1,\ldots,k\); \(N\) – number of experiments – groups of measurements data \((x_{1i}, x_{2i}, \ldots, x_{ki}, y_i), i = 1,\ldots,N; N \geq k; \varphi_j(X) - \) specified system of many variables, independent linearly

Let \(C\) be the vector of parameters values in the \(i\)-th stage of adaptive method. Let assume also, that we obtain a new group of data \(I_{i+1} = (X_{i+1}, Y_{i+1})\). These data should help in replacing \(C_i\) by \(C_{i+1}\), so will be the source of correction:

\[C_{i+1} = C_i + \Delta C_{i+1} \tag{9}\]

Now we will replace \(C_i\) by \(C_{i+1}\). For the local discrepancy between value predicted by model and registered at the station we have:

\[q_{i+1}(C_i) = f(X_{i+1}, C_i) - y_{i+1} \tag{10}\]
the value $\Delta C_{i+1}$ should be such to decrease square of this difference. It is obtained by „antigradient” method, so:

$$\Delta C_{i+1} = -\alpha_{i+1} \nabla q^2(C_i)$$

(11)

where: $\alpha_{i+1}$ – positive coefficient, $\nabla$ - nabla operator, $\nabla = \left( \frac{\partial q}{\partial x}, ..., \frac{\partial q}{\partial k} \right)$. 

$$\nabla q^2(C) = \left( \frac{\partial^2 q}{\partial i^2} \frac{\partial q}{\partial x}, ..., \frac{\partial^2 q}{\partial k^2} \right) = 2 \partial q(C) \left( \frac{\partial q}{\partial i}, ..., \frac{\partial q}{\partial k} \right) = 2 \partial q(C) \nabla q(C)$$

(12)

After transformations, for the function, given by equation (5) we obtain:

$$q = c_0 + c_i \varphi_i(X_i^{i+1}) + ... + c_k \varphi_k(X_k^{i+1}) - y_{i+1}$$
oraz

$$\nabla f(X,C) = \left[ \varphi_1(X_i^{i+1}), ..., \varphi_k(X_k^{i+1}) \right]$$

so:

$$(c_0 - 2ac_1 + 1) + (c_1 - 2ac_1 \varphi_1(X_i^{i+1}) + \varphi_i(X_i^{i+1}) + ... + (c_k - 2ac_k \varphi_k(X_i^{i+1}) + \varphi_k(X_i^{i+1}) - y_{i+1} = 0$$

Solution gives an optimal value of $\alpha^*$, for which the discrepancy $q(C_i + \Delta C_i)$ heads to 0.

$$\alpha^* = \frac{1}{2} \left[ I + \sum_{j=1}^{k} c_j \varphi_j^2(X^{i+1}) \right]^{-1}$$

(13)

where: $X^{i+1}$ - is a system of co-ordinates of start points vector $X$ in the moment $i+1$.

These modelling methods were applied to the data of air pollution and meteorological conditions from the automatic measurement station in Katowice. These data were made available for us by Provincial Inspectorate of Environment Protection in Katowice.

3 Modelling results

According to the rules of ARX models determination, the significance of coefficients in equation (7) was checked by using STATISTICA program, with the displacements (for $p$, $k$, $l$, $r = 0, 1, 2$) for the data from measurement station in Katowice.

The model for heating season 1996/1997 ($R = 0.8519; s_r = 27.66$):

$$SO_2(t) = 55.52 - 10.02v(t) - 2.78T(t-1) + 0.59SO_2(t-1)$$

[6.69] [3.61] [0.42] [0.05]

(in square brackets – standard errors of coefficients, $s_r$ – rest deviation)

The model for heating season 1997/1998 ($R = 0.8055; s_r = 20.43$):

$$SO_2(t) = 37.73 - 9.73v(t) - 1.79T(t-1) + 0.62SO_2(t-1)$$

[4.70] [2.65] [0.33] [0.05]

It occurs from the analysis of equations and their coefficients significance that the obtained regressive equations for the seasons of measurement station in Katowice are very significant and the estimation errors (mean rest deviations) are not large. There is clear difference between the influence of various factors on the $SO_2(t)$ in individual seasons.
The obtained model for heating season 1996/1997 of measurement station in Katowice was the base to determination of coefficients in adaptive model for heating season 1997/1998. In the purpose of avoiding the strong influence of large fluctuations of individual variables, the additional cases were introduced to the adaptive modelling, which were the mean values of each following pairs of days. After calculations it occurred, that the rest deviation for adaptive model amounted 14.81 µg/m³ (calculated for real days) and applying the regressive model from the season 1996/1997 - 22.78 µg/m³. Figure 3 presents course of conformity of SO₂ air pollution prediction in heating season 1997/1998 for adaptive model and for regressive model from season 1996/1997.

![Figure 3: Course of conformity of SO2 air pollution prediction for adaptive regressive models.](image)

### 4 Conclusions

The unfavourable conditions of ventilation occur very often during winter. They are the result of the chill of lower atmosphere’s layers phenomenon, which lead to creation of temperature’s inversion. It limits considerably the vertical exchange of air masses. In effect the pollution introduced to atmosphere from the low emission sources cumulate in its lower layer and there is an increase of pollution concentration near the surface of earth. Furthermore, it is usually connected with weak winds. It is also the factor, which worsen the pollution’s dispersion conditions, so it influences on the quality of air. The chill of the lower atmosphere’s layer is condusive to water vapour condensation – fogs and mists occur very often. In connection with a large content of SO₂ and dusts in air it may lead even to creation of acid smog (winter smog).
The warnings about unfavourable conditions of air pollution are significant actions, because of its harmful influence on people’s health state. So, it can be said, that the correct forecasting is crucial for preventive purposes.

Looking at the results presented above it seems to be correct to conduct these forecasts in the base of adaptive models. Thanks to them it is possible to adjust the model parameters to the change of the factors, which condition the air pollution state.

Acknowledgement

The paper is the effect of statutory work no. 11.11.100.238

References
