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MODELS OF DISTRIBUTION OF POLLUTANTS IN THE AIR

Distribution of pollutants which are emitted from instant and continuous sources is described. This description was obtained on the basis of the theory of partial differential equations of parabolic or elliptical type. This paper also presents a simple way of obtaining analytical solutions. This way consists in using the idea of source function.

System of calculation was examined by comparison of measured concentrations by monitoring net in the area of the chemical factory 'Police' and calculated concentrations of water-soluble compounds of fluorine.

1. INTRODUCTION

Toxic compounds are emitted into the air by point sources, linear sources and surface sources. Spreading of pollutants on a regional or smaller scale is usually described using diffusion models. Diffusion models are partial differential equations of parabolic or elliptical type. On the basis of these equations [1], propagation of the pollutants from point sources was described. These sources can be temporarily or continually active. Both cases were examined.

2. TEMPORAL SOURCES

Extending of pollutants emitted from moment sources is usually described by parabolic equation:

$$\frac{\partial u}{\partial t} = \frac{\partial}{\partial x} \left[D(x) \frac{\partial u}{\partial x} \right] + \frac{\partial}{\partial y} \left[D(y) \frac{\partial u}{\partial y} \right] + \frac{\partial}{\partial z} \left[D(z) \frac{\partial u}{\partial z} \right] + f(x, y, z, t) \quad (1)$$

where

u – concentration of pollutant in the air, g/m^3 ,

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t – time, s,

x, y, z – coordinates of the Cartesian system, m,

$f(x, y, z, t)$ – density of sources of mass, $\text{g/m}^3\text{s}$,

$D(x), D(y), D(z)$ – coefficients of diffusion, m^2/s .

If a medium is homogeneous, then $D(x) = D(y) = D(z) = D$. In the case of various constant coefficients for coordinates, we have got D_1, D_2, D_3 . If density of sources of mass is zero, then we will obtain homogeneous equation

$$u_t = \alpha_1^2 u_{xx} + \alpha_2^2 u_{yy} + \alpha_3^2 u_{zz}, \quad \alpha_1^2 = D_1; \quad \alpha_2^2 = D_2; \quad \alpha_3^2 = D_3. \quad (2)$$

When we want to get a unique solution of diffusion equation, we have to take account of boundary conditions. For parabolic equation, the initial condition is given by the value of the function $u(x, y, z, t)$ for $t = 0$. Boundary conditions can be different, depending on the distribution of concentration at the boundaries of the area. In general, we consider the following types of boundary conditions:

the concentration is given at the boundary of area $\mu(t)$,

the value of differential $\partial u/\partial x$ is given,

linear relationship between differential and function is given.

If we know differential equation and boundary conditions, we will be able to find solution of $u(x, y, z, t)$. In order to contract the notation, we will examine problem with only one spatial variable. In this case, we arrive at

$$u_t = \alpha_1^2 u_{xx} + f(x, t). \quad (3)$$

We assure the initial condition in the following form

$$u(x, 0) = 0 \quad (4)$$

and boundary conditions

$$u(0, t) = u(l, t) = 0. \quad (5)$$

The solution of this problem for the interval l and the time of source activity t can be expressed as

$$\begin{aligned} u(x, t) &= \int_0^t \int_0^l \left\{ \frac{2}{l} \sum_{n=1}^{\infty} \exp \left[- \left(\frac{\pi n}{l} \right)^2 \alpha_1 (t - \tau) \right] \sin \frac{\pi n}{l} x \sin \frac{\pi n}{l} \zeta \right\} f(\zeta, \tau) d\zeta d\tau \\ &= \int_0^t \int_0^l G(x, \zeta, t - \tau) f(\zeta, \tau) d\zeta d\tau. \end{aligned} \quad (6)$$

The expression $G(x, t, \zeta)$, called the source function, is examined as the function x . This function represents the distribution of the concentration of diffused component

the interval $0 \leq x \leq 1$ in the moment t if the concentration at the beginning is zero and at this moment at the point $x = \zeta$ elementary mass of this component is emitted and at the ends of the interval the zero concentrations are kept. For every $0 \leq x \leq 1$ and for every $t > 0$, the function $G(x, t, \zeta) \geq 0$.

If the power of source is Q_0 , then

$$u(x, t) = Q_0 G(x, t, \zeta). \quad (7)$$

Hence, the solution of the problem of diffusion is reached, provided that we know how to find the function of the source $G(x, t, \zeta)$.

If pollutants are extending the air for coordinate x we are interested in the function of the source for infinite line. This function for coordinate x has the following form:

$$G(x, t, \zeta) = \frac{1}{2\sqrt{\pi\alpha_1^2 t}} \exp\left[-\frac{(x-\zeta)^2}{4\alpha_1^2 t}\right]. \quad (8)$$

The dependence (8), expressed by Gauss' function, can be named the basic solution of the diffusion equation.

Analogously, the function of the source for coordinate y is expressed as:

$$G(y, t, \eta) = \frac{1}{2\sqrt{\pi\alpha_2^2 t}} \exp\left[-\frac{(y-\eta)^2}{4\alpha_2^2 t}\right]. \quad (9)$$

Construction of the function of the source for coordinate z (height) is little different. Coordinate z changes in the range $0 \leq z \leq \infty$. Therefore, we ought to search for the solution of diffusion equation for half-line. Effective heights of sources of emission are relatively small compared to the range of changes of coordinate z . For this reason we have to examine the case where the source is close to the origin of the half-line. So we search for the solution of the diffusion equation for $t > 0$ at the initial condition

$$u(z, 0) = \varphi(z) \quad (10)$$

and one of the three boundary conditions which are dependent on the character of the process. In the theory of propagation of pollutants in the air, we usually examine the second boundary condition $\partial u(0, t) / \partial z = 0$.

The function of the source at homogeneous boundary condition of the second order and the initial condition (10) is described as follows:

$$G(z, t, H) = \frac{1}{2\sqrt{\pi\alpha_3^2 t}} \left\{ \exp\left[-\frac{(z-H)^2}{4\alpha_3^2 t}\right] + \exp\left[-\frac{(z+H)^2}{4\alpha_3^2 t}\right] \right\} \quad (11)$$

where H is effective height. The method of calculation of this parameter was described in [2].

Based on the theory of parabolic equations [1] we are able to describe the function of the source for three-dimensional space:

$$G(x,y,z,t; \zeta, \eta, H) = G_x G_y G_z = \left(\frac{1}{2\sqrt{\pi t}} \right)^3 \frac{1}{\alpha_1 \alpha_2 \alpha_3} \exp \left[- \frac{(x - \zeta)^2}{4\alpha_1^2 t} \right] \\ \times \exp \left[- \frac{(y - \eta)^2}{4\alpha_2^2 t} \right] \left\{ \exp \left[- \frac{(z - H)^2}{4\alpha_3^2 t} \right] + \exp \left[- \frac{(z + H)^2}{4\alpha_3^2 t} \right] \right\}. \quad (12)$$

Equation (12) is valid if the medium is immovable, i.e. the position of a centre of the cloud emitted is constant when the cloud is extending. If the masses of the air move in time along to coordinate x with mean speed $|\vartheta_1|$, then the position of a centre of the cloud changes according to the equation

$$\zeta = |\vartheta| t. \quad (13)$$

Coefficients $\alpha_1, \alpha_2, \alpha_3$ should be determined experimentally.

3. DIFFUSION IN THE FLUX AND CONTINUOUS SOURCES

Let us consider diffusion of one component in nonstationary flux without sources. For this case, we arrive at the following diffusion equation:

$$\frac{\partial u}{\partial t} = \frac{\partial}{\partial x} \left[D(x) \frac{\partial u}{\partial x} \right] + \frac{\partial}{\partial y} \left[D(y) \frac{\partial u}{\partial y} \right] + \frac{\partial}{\partial z} \left[D(z) \frac{\partial u}{\partial z} \right] - \frac{\partial}{\partial x} (\vartheta_1 u) - \frac{\partial}{\partial y} (\vartheta_2 u) - \frac{\partial}{\partial z} (\vartheta_3 u). \quad (14)$$

If we assure that in the space $z \geq 0$, the flux is directed along the coordinate x and moves with a mean speed $|\vartheta_1|$, then equation (14) is reduced to:

$$\frac{\partial u}{\partial t} = \frac{\partial}{\partial x} \left[D(x) \frac{\partial u}{\partial x} \right] + \frac{\partial}{\partial y} \left[D(y) \frac{\partial u}{\partial y} \right] + \frac{\partial}{\partial z} \left[D(z) \frac{\partial u}{\partial z} \right] - |\vartheta_1| \frac{\partial u}{\partial x}. \quad (15)$$

Due to construction of function of the source $G = G_x G_y G_z$ we arrive at the solution of equation (15). Function of source G_x is obtained as a result of solving the problem for the straight line x , provided that $D(x) = D_1$. For this case, the diffusion equation takes the form

$$\frac{\partial u}{\partial t} = \alpha_1^2 \frac{\partial^2 u}{\partial x^2} - |\vartheta_1| \frac{\partial u}{\partial x}, \quad \alpha_1^2 = D_1, \quad -\infty < x < \infty. \quad (16)$$

Inserting $\theta = \alpha_1^2 t$ and $\beta = |\vartheta_1|/\alpha_1^2$ we obtain:

$$\frac{\partial u}{\partial \theta} = \frac{\partial^2 u}{\partial x^2} - \beta \frac{\partial u}{\partial x}. \quad (17)$$

Thereafter we insert

$$u = ve^{\mu x}. \quad (18)$$

and calculate the differentials $\partial u/\partial \theta$, $\partial u/\partial x$, and $\partial^2 u/\partial x^2$. The result is inserted into (18) and we arrive at

$$\frac{\partial v}{\partial \theta} = \frac{\partial^2 v}{\partial x^2} - \mu^2 v = \frac{\partial^2 v}{\partial x^2} + cv \quad (19)$$

where

$$c = -\mu^2 = \left(\frac{1}{2} \frac{|\vartheta_1|}{\alpha_1^2} \right)^2.$$

We solve equation (19) by substituting

$$v = \tilde{v}e^{c\theta} \quad (20)$$

Then we calculate the differentials $\partial \tilde{v}/\partial \theta$ and $\partial^2 \tilde{v}/\partial x^2$ and introduce them to (19). As a result we obtain

$$\frac{\partial \tilde{v}}{\partial \theta} = \frac{\partial^2 \tilde{v}}{\partial x^2} \quad (21)$$

or

$$\frac{\partial \tilde{v}}{\partial \theta} = \alpha_1^2 \frac{\partial^2 \tilde{v}}{\partial x^2}. \quad (21')$$

The respective functions of the source for equations (21'), (17) and (16) are as follows:

$$\tilde{G}_v = \frac{1}{2\sqrt{\pi\alpha_1^2 t}} \exp\left[-\frac{x^2}{4\alpha_1^2 t}\right], \quad (22)$$

$$G_v = \tilde{G}_v e^{c\theta} = \frac{1}{2\sqrt{\pi\alpha_1^2 t}} \exp\left[-\frac{x^2}{4\alpha_1^2 t} - \frac{1}{4} \left(\frac{|\vartheta_1|}{\alpha_1}\right)^2 t\right], \quad (23)$$

$$G_x = G_v e^{\mu x} = \frac{1}{2\sqrt{\pi\alpha_1^2 t}} \exp \left[-\frac{(x - |\vartheta_1| t)^2}{4\alpha_1^2 t} \right]. \quad (24)$$

Function of the source G_y derived on the basis of the following equation

$$|\vartheta_1| \frac{\partial u}{\partial x} = \alpha_2^2 \frac{\partial u^2}{\partial y^2}, \quad -\infty < y < \infty \quad (25)$$

takes the form

$$G_y = \frac{1}{2\sqrt{\pi\bar{\sigma}_y}} \exp \left[-\frac{(y - \eta)^2}{4\bar{\sigma}_y^2} \right] \quad (26)$$

where

$$\bar{\sigma}_y^2 = \frac{D_2}{|\vartheta_1|} x$$

denotes a coefficient.

Function of the source G_z is defined based on the equation similar to (25). This equation is defined for coordinate z . We can solve it at $z \geq 0$ and the second boundary condition $\partial(0, t)/\partial z = 0$ which gives

$$G_z = \frac{1}{2\sqrt{\pi\bar{\sigma}_z}} \left\{ \exp \left[-\frac{(z - H)^2}{4\bar{\sigma}_z^2} \right] + \exp \left[-\frac{(z + H)^2}{4\bar{\sigma}_z^2} \right] \right\} \quad (27)$$

where

$$\bar{\sigma}_z^2 = \frac{D_3}{|\vartheta_1|} x$$

denotes a coefficient.

The coefficients $\bar{\sigma}_y$, $\bar{\sigma}_z$ are expressed in meters and calculated experimentally. Function of the source for three-dimensional space can be obtained from the relationship

$$G(x, y, z, t; \vartheta_1, \eta, H) = G_x G_y G_z. \quad (28)$$

If function of the source is known, we arrive at the equation for temporal-spatial distribution of concentration in nonstationary flux of air

$$u(x, y, z, t) = \left(\frac{1}{2\sqrt{\pi}}\right)^3 \frac{Q}{\bar{\sigma}_x \bar{\sigma}_y \bar{\sigma}_z} \exp\left[-\frac{(x - |\vartheta_1|t)^2}{4\bar{\sigma}_x^2}\right] \exp\left[-\frac{(y - \eta)^2}{4\bar{\sigma}_y^2}\right] \\ \times \left\{ \exp\left[-\frac{(z - H)^2}{4\bar{\sigma}_z^2}\right] + \exp\left[-\frac{(z + H)^2}{4\bar{\sigma}_z^2}\right] \right\} \quad (29)$$

where

$$\bar{\sigma}_x^2 = \alpha_1^2 t, \text{ m,}$$

Q – amount of the substance emitted, kg.

The equation, which describes a spatial distribution of concentration in the flux for point continuous source ($t \rightarrow \infty$) of constant output $\varphi(t) = \dot{Q}$, is obtained from nonhomogeneous solution of diffusion equation:

$$\bar{u}(x, y, z) = \int_0^{\infty} G_x G_y G_z \varphi(t) dt = \dot{Q} G_y G_z \int_0^{\infty} G_x dt = \frac{\dot{Q}}{4\pi \bar{\sigma}_y \bar{\sigma}_z |\vartheta_1|} \exp\left[-\frac{(y - \eta)^2}{4\bar{\sigma}_y^2}\right] \\ \times \left\{ \exp\left[-\frac{(z - H)^2}{4\bar{\sigma}_z^2}\right] + \exp\left[-\frac{(z + H)^2}{4\bar{\sigma}_z^2}\right] \right\} \quad (30)$$

where \dot{Q} denotes a flux of mass emitted, kg/s. In calculations, we usually assure that \dot{Q} is a mean annual concentration.

We arrive at equations (29) and (30) assuming that the pollutants emitted are not transformed in the air, air masses move along x axis with average velocity $|\vartheta_1|$, coordinates change in the ranges $-\infty < x < \infty$, $-\infty < y < \infty$, $z \geq 0$ and the second boundary condition is fulfilled. So equations (29), (30) are valid for these conditions only.

The description presented formally can be considered as Euler's model. Especially this is a proposition of obtaining analytical solutions.

$\bar{\sigma}_y$, $\bar{\sigma}_z$ are coefficients of the model. They are usually described by empirical equations [3], [4].

Comparison of the equations obtained with the Gauss model is interesting. From mathematical description of final equations for both models it follows that application of the equations allows us to obtain the same results, provided that numerical values of the coefficient fulfil the following relations:

$$\sigma_y = \sqrt{2} \bar{\sigma}_y, \quad (31)$$

$$\sigma_z = \sqrt{2} \bar{\sigma}_z. \quad (32)$$

Verification is necessary for one model only, because the second will also be valid if relations (31) and (32) are taken into account.

4. SYSTEMS OF EVALUATION OF AIR QUALITY

According to general standards, the quality of air is estimated on the basis of instantaneous, average twenty-four hours and annual average concentrations [5], [6]. In general, only instantaneous and annual average concentrations are determined [7]. They can be obtained experimentally or calculated using adequate mathematical models.

Calculation of distribution of concentrations requires adequate systems. These systems contain equations describing temporal-spatial distributions of concentrations, correlations determining the parameters of these equations and adequate method of taking account of meteorological conditions. We know various systems, for example, Jülich's, Pasquille's, Brigs', St. Luiss', Klugs' systems [8]. The parameters σ_x , σ_y , σ_z are calculated from empirical equations; the parameters σ_x , σ_y are usually described by the equation

$$\sigma_x = \sigma_y = Ax^a \quad (33)$$

where

x – distance from a source, m,
 A , a – empirical coefficients.

More types of relations are used to describe the parameter σ_z [8]. The well-known relations can be itemized as follows:

Pasquille's, Gifford's, Dingler's, Smith's (PGSS) and Klug's equation

$$\sigma_z = ax^b, \quad (34)$$

Martin's–Tikvart's equation

$$\sigma_z = ax^b + c, \quad (35)$$

Brigs' equation

$$\sigma_z = ax(1 + bx)^c \quad (36)$$

where

x – distance from a source, m,
 a , b , c – empirical coefficients.

The guidelines laid down in Poland are based on Pasquille's system [7]. Equations of concentration distribution are as follows:

$$u(x, y, z, t) = \left(\frac{1}{2\sqrt{\pi}} \right)^3 \frac{Q}{\sigma_x \sigma_y \sigma_z} \exp \left[-\frac{(x - |\vartheta_1|t)^2}{2\sigma_x^2} \right] \exp \left[-\frac{(y - \eta)^2}{2\sigma_y^2} \right] \\ \times \left\{ \exp \left[-\frac{(z - H)^2}{2\sigma_z^2} \right] + \exp \left[-\frac{(z + H)^2}{2\sigma_z^2} \right] \right\} \quad [\text{g/m}^3], \quad (37)$$

for annual average concentration

$$\bar{u}(x, y, z) = \int_0^{\infty} u(x, y, z, t) dt = \frac{\dot{Q}}{2\pi\sigma_y\sigma_z|\vartheta_1|} \exp \left[-\frac{(y - \eta)^2}{2\sigma_y^2} \right] \\ \times \left\{ \exp \left[-\frac{(z - H)^2}{2\sigma_z^2} \right] + \exp \left[-\frac{(z + H)^2}{2\sigma_z^2} \right] \right\} \quad [\text{g/m}^3]. \quad (38)$$

In the guidelines, the parameters in equations (37), (38) are described as follows:

$$\sigma_x = \sigma_y = Ax^a, \quad (39)$$

$$\sigma_z = Bx^b \quad (40)$$

where

$$a = 0.367 (-2.5 m),$$

$$b = 1.55 \exp(-2.35 m),$$

$$A = 0.08 [6m^{-0.3} + 1 - \ln(H/z_0)],$$

$$B = 0.38 m^{1.3} [8.7 - \ln(H/z_0)],$$

m – meteorological parameter (data from table),

H – effective height of emitter,

z_0 – correction coefficient for area investigated (data from table).

Before measurements of immission concentrations we should verify the method applied to the given area, which is done by comparing the concentrations calculated and measured. We assume that model substance is emitted from only one object, it cannot be transferred from other areas, because it is difficult to estimate the share of the substance transferred in total concentration. It should be stressed that consistency of the concentrations measured and calculated depends not only on the mathematical model but also on the accuracy of measurements done at particular points of monitoring net and on the accuracy of emission estimation.

Such a verification of measurements was carried out for area of Szczecin in 1992 and 1993. Water-soluble fluorine compounds were assumed to be model substances because they were emitted by one factory. Distributions of instantaneous concentra-

tions S_{30} and annual average concentrations S_a of fluorine compounds in the air were calculated based on the emission quantity determined experimentally and technical parameters of emitters. The data for 1993 is given in table 1.

Table 1

Characteristics of sources of emission of fluorine compounds

Symbol of emitter	X_e [m]	Y_e [m]	Emission of fluorine compounds [kg/h]	d [m]	V_s [m/s]	T [K]	H [m]
Po-1	0	0	0.0645	1.20	10.5	333	45
Po-2	80	-40	0.0657	0.60	19.5	313	25
Po-3	145	60	0.1200	1.20	10.5	333	45
Po-4	165	10	0.1363	0.60	19.5	313	25
Po-5	10	120	0.8300	0.31	5.8	311	17
Po-6	265	120	0.3404	1.60	13.8	340	45
Po-7	290	50	0.0680	0.64	12.7	325	32
Po-8	-20	-190	0.2670	3.96	11.9	328	60
Po-9	215	-105	2.7412	3.96	12.4	328	60
Po-10	-575	-515	0.1524	3.20	10.2	303	42
Po-11	-575	-515	0.2836	4.20	6.0	318	42

Figures 1 and 2 present the chosen isolines of annual average concentration of fluorine compounds. They also show the position of points of monitoring net. Calculated values of instantaneous and annual average concentrations were taken from these figures for each point of monitoring net and gathered in tables 2 and 3.

Table 2

Comparison of the calculated and measured concentrations of water-soluble fluorine compounds, area of Police, 1992

No.	Locality	Model concentration		Measured concentration	
		S_{30} [$\mu\text{g}/\text{m}^3$]	S_a [$\mu\text{g}/\text{m}^3$]	S_{ap} [$\mu\text{g}/\text{m}^3$]	S_{ap}/S_a
1	Jasienica	8	0.08	0.2	2.5
2	Tatynia	7	0.11	0.1	0.9
3	Police-Fabryczna	9	0.13	0.2	1.5
4	Police-Stadion	8	0.12	0.2	1.7
5	Police-Hotel	8	0.09	0.2	2.2
6	Wieńkowo	7	0.11	0.2	1.8
average 1.8					

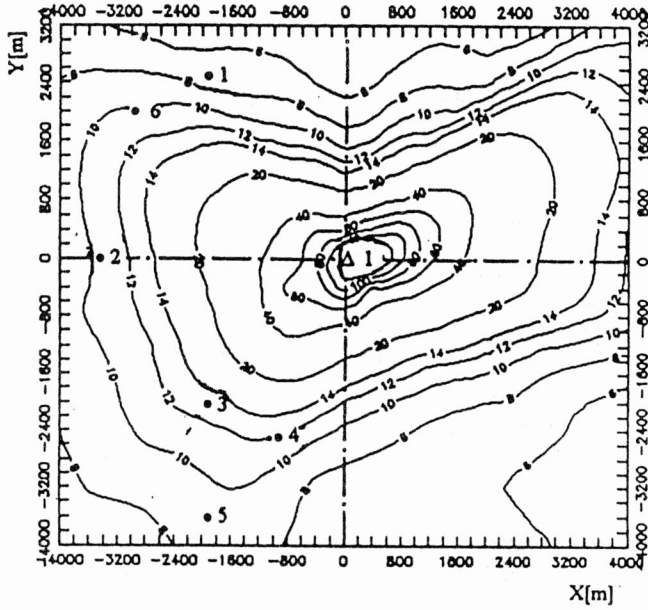


Fig. 1. Chosen isolines of annual average concentration $S_a \cdot 100$ [$\mu\text{g}/\text{m}^3$] of water-soluble fluorine compounds. Area of Police, 1992

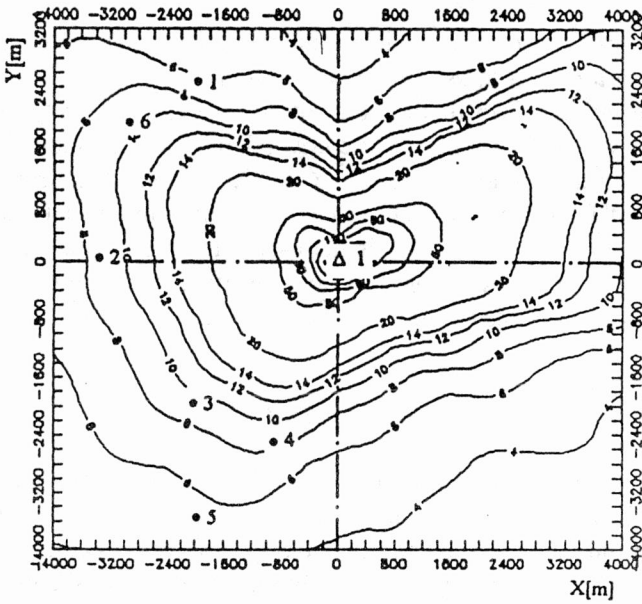


Fig. 2. Chosen isolines of annual average concentration $S_a \cdot 100$ [$\mu\text{g}/\text{m}^3$] of water-soluble fluorine compounds. Area of Police, 1993

Table 3

Comparison of the calculated and measured concentrations of water-soluble fluorine compounds, area of Police, 1993

No.	Locality	Model concentration		Measured concentration	
		S_{30} [$\mu\text{g}/\text{m}^3$]	S_a [$\mu\text{g}/\text{m}^3$]	S_{ap} [$\mu\text{g}/\text{m}^3$]	S_{ap}/S_a
1	Jasienica	11	0.07	0.1	1.4
2	Tatynia	10	0.09	0.1	1.1
3	Police-Fabryczna	11	0.09	0.2	2.2
4	Police-Stadion	11	0.09	0.1	1.1
5	Police-Hotel	8	0.06	0.2	3.3
6	Wieńkowo	10	0.10	0.1	1.0

average 1.7

Measured values of annual average concentrations in each measuring position were based on twenty-four hour continuous measurements. Therefore, at the beginning, average concentrations for each month were calculated, and next – for each year (tables 2 and 3).

We stated strict conformity of the results in some measuring positions but in certain positions subjected to investigations in 1992–1993 discrepancy of the results was wide.

Small number of final results, which results from the number of points of monitoring net, does not allow us to use statistical methods.

Average value of the ratio of the concentration measured and calculated is 1.8, which proves that we deal with systematic error. This error can result from the model or manner of determining annual average concentrations (accuracy of analytical method, number of measurements, etc.). There are no simple methods to solve this problem. We can only investigate simultaneously some model substances in the same area [9]. According to this estimation, this error results from the method of calculating annual average concentrations of fluorine compounds.

5. SUMMARY

Basic models used to describe distribution of pollutants in the air are as follows: differential models of Euler, integral models of Lagrange and models of Gauss.

Using differential models in the form of partial differential equations of parabolic and elliptic types we may present the distribution of pollutants emitted from point, continuous and instantaneous sources. We describe a simple way of obtaining the analytical solutions which seem to be important in theoretical considerations. This way consists in application of the idea of function of the source. The manner of cal-

ulation was verified in 1992–1993 by comparison of the calculated and measured annual average concentrations of water-soluble fluorine compounds from monitoring net in the area of chemical factory 'Police'.

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MODELE ROZPRZESTRZENIANIA ZANIECZYSZCZEŃ W ATMOSFERZE

Substancje toksyczne są wyrzucane do atmosfery przez źródła punktowe, liniowe bądź powierzchniowe. Rozprzestrzenianie zanieczyszczeń w skali regionalnej lub mniejszej zwykle opisuje się stosując modele dyfuzyjne. Stanowią je cząstkowe równania różniczkowe typu parabolicznego albo eliptycznego. W pracy, na podstawie teorii tych równań [1], opisano propagację zanieczyszczeń ze źródeł punktowych. Źródła takie mogą działać chwilowo albo ciągle. Rozpatrzono oba przypadki.

