

RISKS POSED BY PARTICULATE MATTER TO THE HUMAN HEALTH AND ENVIRONMENT NEAR TRANSPORT ROUTES

ZDZISŁAW CHŁOPEK¹, KATARZYNA SUCHOCKA²

Warsaw University of Technology, Automotive Industry Institute (PIMOT)

Summary

Dusts pose serious danger to the human health and environment. A significant part of dust emissions³ comes from motor traffic. In consideration of the very harmful impact of dusts, increasingly restrictive limitations are imposed by law on the dust immissions⁴ in atmospheric air. Originally, the immission of particulate matter fraction PM10 (with characteristic particle dimension below 10 μm) was limited by law. Since 2010, the immission limitations have been extended to the particulate matter fraction PM2.5 (with characteristic particle dimension below 2.5 μm). The environmental risks posed by dusts are assessed on the grounds of measurements, modelling of pollutant emissions from natural sources and from sources connected with civilization, and modelling of dissemination of such pollutants. This paper presents models of the immission of specific size fractions of the particulate matter coming from automotive sources, based on functional similarity (behaviouristic models). In the PM10 immission models, a relationship is postulated according to which the PM10 particulate matter immission rises with increasing nitrogen oxides and carbon monoxide immissions. In the PM2.5 particulate matter immission models, an increasing dependence of the PM2.5 immission on the PM10 immission is postulated. Similarly, the PM1 immission is postulated to increase with rising PM2.5 and PM10 particulate matter immissions. The paper shows results of identification of the PM10 and PM2.5 immission models for two air quality monitoring stations. The model coefficients were found to be very susceptible to the conditions of pollutant emission (especially to the types of pollutant emission sources) and to the pollutant dissemination conditions.

Keywords: pollutant emissions, particulate matter, motorization

1. Introduction

Dust is one of the pollutants that are most harmful to the environment. This was pointed out as long ago as in 1524 by Georgius Agricola, who wrote about the harmful impact of dust on the human health in his work "De re metalica" [1]. Especially in urban agglomerations, particulate matter fractions PM10 and PM2.5 are counted among the pollutants the

¹ Warsaw University of Technology, Institute of Vehicles, ul. Narbutta 84, 02-524 Warsaw, Poland, e-mail: zchlopek@simr.pw.edu.pl, ph. +48 22 234 85 59

² Automotive Industry Institute, Environmental Protection and Natural Energy Use Department, ul. Jagiellońska 55, 03-301 Warsaw, Poland, e-mail: k.suchocka@pimot.eu

³ Pursuant to the "Environmental Protection" Act of 27 April 2001, the term "emission" as a phenomenon means the environmental impact exerted by introducing to the environment substances or energies recognized as environmentally harmful. As a physical quantity, "pollutant emission" is defined as the mass of a pollutant introduced to the environment [1, 24, 28].

⁴ „Immission" is the concentration of a pollutant dispersed in the atmospheric air, measured at a height of 1.5 m above the Earth's surface [21].

immissions of which most often exceed the permissible limits [2, 4, 8–11, 14, 15, 17, 25]. Dusts are composed of particles having different dimensions. The term "dust" is defined as the dispersed phase of a two-phase system consisting of solid body, i.e. small solid particles, suspended in gaseous dispersion medium, i.e. air [21, 32]. In general, the shape of the solid particles (or grains) that constitute dust is not spherical. Therefore, the idea of equivalent particle sizes has been adopted for evaluating the characteristic particle dimensions. There are many methods of defining the equivalent particle sizes. Such methods chiefly depend on the measurement techniques used, which are specified in the relevant regulations [3, 16, 29–31]. Usually, dust particles are considered in relation to the comparable spherical grains. For this reason, the term "equivalent diameter" or "equivalent size" will be used throughout this paper.

Depending on the conventional dust particle size, the following dust categories are defined [3, 16, 18, 29–31]:

- TSP (total suspended particles), with equivalent particle size below 300 μm ;
- Fine-grained dust, consisting of particles of equivalent size below 75 μm , which settle out under their own weight but may remain suspended for some time;
- Fine dust PM₁₀, with equivalent particle size below 10 μm ;
- Fine dust PM_{2.5}, with equivalent particle size below 2.5 μm ;
- Dust PM₁, with equivalent particle size below 1 μm ;
- Nanoparticles⁵, with equivalent particle size below 100 nm [24, 28] (dust invisible to the naked eye).

The particulate matter fractions PM₁₀ and PM_{2.5} are defined in the standards where the method of measuring particle dimensions is specified. The particulate matter PM₁₀ is the dust that can go through a size-selective inlet as defined in the reference method for the sampling and measurement of PM₁₀ (see EN 12341) with a 50% efficiency cut-off at an aerodynamic diameter (AED) of the particle of up to 10 μm [16, 30, 31]. The particulate matter PM_{2.5} is the dust that can go through a size-selective inlet as defined in the reference method for the sampling and measurement of PM_{2.5} (see EN 14907) with a 50% efficiency cut-off at an aerodynamic diameter (AED) of the particle of up to 2.5 μm [16, 30, 31]. Similar definitions are applicable to other particle size fractions, e.g. PM₁.

Apart from the above, a term "respirable dust" can also be met; moreover, dust particles are sometimes classified depending on the depth of their penetration into the human respiratory tract [16, 18, 30, 31].

The respirable dust is a set of the particles that can pass through a preliminary particle selector with size-permeability (based on equivalent particle sizes) described by a log-normal probability function with an average AED value of $3.5 \pm 0.3 \mu\text{m}$ and a standard deviation of $1.5 \pm 0.1 \mu\text{m}$.

According to standard EN 481 [16], two dust fractions are defined:

- Inhalable fraction (that can be breathed into the nose or mouth);
- Thoracic fraction (that can penetrate to the bronchioles).

⁵ There are some differences in defining the term "nanoparticles". This is, to a significant extent, because of the spontaneous introduction of this idea both in the field of sciences and in non-scientific activities and due to a tendency observed for the recent years that commercial expressions are readily adopted by scientific circles to function as scientific terms. So, the values of 50 nm or even 1 μm can be met as the upper limit defining the equivalent size of nanoparticles [24, 28].

The dust penetration into the respiratory tract depends on dust particle dimensions [18, 22, 25, 30, 32]. The inhalable fraction consists of particles with equivalent sizes smaller than 100 μm . Particles with dimensions bigger than 30 μm are arrested in the upper part of the respiratory tract (nose, mouth, throat, larynx) and then excreted with mucus. The middle parts of the respiratory tract (trachea, bronchi, bronchioles) are reached by the thoracic fraction, the particle size of which does not exceed 20 μm . These particles may accumulate in the upper and middle parts of the respiratory tract. The gas-exchange regions (alveoli) are reached by the particles whose dimensions are smaller than 7 μm . Such particles are the basic component of the respirable dust, which remains there for a quite long time causing pathological changes. The particles with dimensions smaller than 2.5 μm penetrate to, and accumulate in, the deepest lung regions. The dust particles soluble in body fluids penetrate directly into the blood.

The harmful impact of dust on the human and animal health depends on the size, chemical and mineralogical composition, and physical structure of dust particles [18, 22, 25, 30, 32]. In general, an opinion is formulated that the most harmful impact on the human health is exerted by the fine and very fine dust particles, which deeply penetrate into the respiratory tract. The particulate matter fractions PM10 and PM2.5 cause various respiratory diseases such as asthma or chronic bronchitis; they are also a factor aggravating the symptoms of chronic obstructive pulmonary disease (COPD) [22]. Particularly harmful to the human health are the dusts that contain particles with heavy metal compounds (especially arsenic, lead, cadmium, nickel, and mercury) and the particles containing heavy cyclic hydrocarbons, as many of the components of such dusts have mutagenic or carcinogenic properties.

Dust exerts harmful impact not only on human and animal health but also on plants, soil, and water. Together with sulphur dioxide, carbon monoxide, and other chemical compounds, dust contributes to the generation of the London smog [9]. Noteworthy is also the fact that dust reduces the visibility, which may be considered a factor that impairs the road safety [9].

Particulate matter emissions result from both natural (non-anthropogenic) and civilization (anthropogenic) processes [32]. The basic natural sources of dust emissions are volcano eruptions, forest fires, wind (eolian) erosion, as well as marine, vegetable and animal aerosols. The anthropogenic causes for the dust emission actually include all the civilization activities, related both to production processes and to the satisfying of people's living needs, especially to the use of the domestic fires where solid fuels are burnt. Among the production processes, chiefly those related to the activities of power industry, cement production industry, building industry, and transport should be mentioned here.

Motor transport is a serious source of dust emissions [2, 4–11, 14, 15, 17, 24, 28, 33]. This is the more so dangerous that the dust emissions related to the use of motor vehicles are particularly high in the central parts of urban agglomerations, where many people, both permanent inhabitants and temporary visitors, are exposed to the air pollution.

The internal combustion (IC) engines, especially those of compression-ignition (CI) type, emit fine particulate matter consisting of carbon base (soot) and organic and

inorganic fractions, which include substances particularly harmful to human health, e.g. heavy organic compounds (such as benzo[a]pyrene, benz[a]anthracene, or benzo[b]fluoranthene) and heavy metals [2, 15, 24, 28, 33]. The particulate matter contained in the IC engine exhaust gases are counted among fine particles, in a significant part having equivalent sizes smaller than 10 μm [24, 28]. The exhaust gases of spark-ignition (SI) engines, especially of those with direct fuel injection, contain very fine particles (referred to as "nanoparticles" [24]); admittedly, the concentration of such particles is rather small, but they are extremely dangerous to human health. For this reason, not only the mass but also the number of such particles is taken into account in the criterion-based methods of testing their emissions.

Other automotive sources of particulate matter emissions include the material stirred up by moving road vehicles from the road surface and the tribological pairs in the vehicle-road system. An important part of these emissions comes from friction pairs in brake mechanisms. Up to 0.5 kg of dust may be annually emitted from the braking system of a passenger car, according to estimates [7]. In addition to this, the solid particles emitted from braking mechanisms are counted among very fine dusts, whose equivalent sizes are of the order of several micrometers or even below 1 μm [19, 20, 26]. The dust emitted from braking mechanisms predominantly consists of iron and iron compounds, chiefly oxides. Other metals, such as barium, magnesium, aluminium, zinc, calcium, copper, silver, molybdenum, antimony, and chromium, are also found to be present in the dust grains [19, 20, 26].

Apart from the emissions of nitrogen oxides, the particulate matter emissions are considered one of the most severe environmental problems caused by the motorization. Therefore, various legal, organizational, and technical measures are taken to reduce the emissions of solid particles. As regards the particulate matter emitted from IC engines, the emission of this pollutant could be radically reduced thanks to new type-approval regulations. For other automotive dust emission sources, no equally effective actions have been recorded so far. In this connection, work to reduce the particulate matter emission from brake mechanisms was undertaken at PIMOT (Automotive Industry Institute). The systems intended for this purpose and preliminarily designed for laboratory tests proved to be very effective, according to rig test results: the coefficient of effectiveness of the reduction of brake dust emission was found to reach a value of up to 0.8 [5–7]. At present, a research and development project No. 10-0050-10/2010 entitled "Development of systems to reduce dust emission from disc brake and drum brake mechanisms of motor vehicles" funded by the National Research and Development Centre is being carried out at PIMOT.

This paper deals with the risks posed by the emissions of solid particles to the environment in large urban agglomerations. This is a difficult problem because continuous monitoring of the immission of specific size fractions of particulate matter is only possible in rather few measuring points, usually at air quality monitoring stations. In the other areas, the environmental risks posed by dusts can only be assessed with employing indirect methods, i.e. with modelling the emission of particulate matter from automotive sources and the dissemination of such pollutants or with modelling the immission of solid particles.

2. Legal situation in the field of environmental protection against the risks posed by particulate matter

In Poland, the common laws being currently effective derive from the following sources: Constitution of the Republic of Poland, Acts of Parliament, ratified international agreements, regulations, and local legal acts.

Originally, the EU legal regulations concerning the protection of people and their environment from dust covered the problem of measuring the immission of total suspended particles; afterwards, this coverage was extended to particulate matter P10. When research results indicated fine particles to be most dangerous for the human health and environment, the introduction of an obligation to measure the PM_{2.5} dust immissions was begun. In the future, obligatory measurements of the PM₁ dust immissions will most likely be introduced; such immissions are already now monitored at some measuring stations. Until 2008, there were four Directives and one Decision that pertained to air quality. Finally, they were combined into one document, i.e. Directive 2008/50/EC of the European Parliament and of the Council of 21 May 2008 on ambient air quality and cleaner air for Europe, sometimes referred to as the CAFE (Clean Air for Europe) Directive.

In this Directive, new obligations of the Member States as regards the monitoring and assessment of the immission of respirable dust PM_{2.5} have been laid down. The Directive formulates six main objectives. The first one is to define and specify the actions concerning ambient air quality and aimed at preventing or reducing the harmful impact of air pollutants on the human health and environment. The second one is to develop common methods and criteria of air quality assessment in all the Member States. The third one is to introduce mutual exchange of information on ambient air quality and to provide mutual help in reducing the emission of pollutants to the atmosphere, with mutual monitoring of long-term trends and improvement in air quality in consequence of that. The fourth objective is to inform the public about the current state of ambient air quality. The fifth one is to maintain the air quality where it is good and to seek for improvements where the pollutant immission limits are exceeded. The sixth objective formulated in the Directive is to promote cooperation between the Member States in the field of reducing the gaseous and dust pollutant emissions to the atmospheric air.

The ambient air quality must be assessed in every zone and agglomeration within the territory of a Member State. If the upper assessment threshold for a pollutant immission is exceeded, fixed measurements should be mandatory. However, the information obtained from fixed measurements may be supplemented by modelling techniques. If the pollutant immission is below the upper assessment threshold then a combination of fixed measurements and modelling techniques and/or measurements referred to as "indicative" may be used. In the case that the pollutant immission is below the lower assessment threshold then modelling or methods referred to as "objective-estimation techniques" alone may be considered sufficient to assess the ambient air quality. For the PM₁₀ dust, the upper and lower pollutant immission assessment thresholds are 28 µg/m³ and 20 µg/m³, respectively. For particulate matter PM_{2.5}, lower values, i.e. 17 µg/m³ and 12 µg/m³, have been adopted for the upper and lower assessment thresholds, respectively.

The EU Member States are obliged to take all the possible measures in order to reduce the risks posed by particulate matter. In particular, the EU Member States shall reduce the PM_{2.5} dust immission to a level below a limit of 20 µg/m³ by 2015.

Annex V to the Directive provides information concerning the criteria for determining the minimum numbers of sampling points for fixed measurements of the immission of gaseous and dust pollutants in the ambient air. The number of the sampling points depends on the population of a specific zone or agglomeration and on the pollutant immissions, i.e. on the question whether the upper assessment threshold is exceeded or not.

The most important Polish document concerning the environmental protection is the "Environmental Protection Law" Act of 27 April 2001. This Act is supplemented with implementing regulations, especially the Regulations of the Minister of the Environment to the Acts of Parliament, e.g. Regulation of 24 August 2012 on the levels of concentration of some substances in the ambient air (Dz. U. of 2012, item 1031).

3. Modelling of the immission of specific particulate matter size fractions

The state of risk posed by specific particulate matter size fractions to the ambient air quality is assessed on the grounds of direct measurements or, where such measurements are not carried out, on the grounds of modelling techniques.

For the environmental risks posed by dusts to be assessed, the immissions of individual particulate matter fractions in the monitored areas must be known. Usually, the immission values are averaged for specific time intervals as required by the applicable legal regulations. Since it is impracticable to measure dust immissions in all the monitored areas, considerable importance is attached to the modelling of particulate matter emissions and dissemination in the environment. The modelling of pollutant emissions makes it possible to assess the state of environmental risk based on analysing the results of measurements of the quantities that are easier to be determined e.g. from inventories of the pollutant emissions from stationary sources and from motor transport sources based on an analysis of traffic intensity and vehicle structure evaluated in terms of the intended use, conventional sizes, and ecological characteristics of the vehicles [4, 10].

The modelling of pollutant dissemination pertains to the relationship between the pollutant immission and the processes that determine the immission, first of all the pollutant emission intensity (rate), which is a derivative of the pollutant emission with respect to time [4, 10]. The relationship between the pollutant immission and the pollutant emission rate in the time domain is of the operational type [4], but the relationship between the average values of the rates of pollutant emission and immission may be treated as a functional dependence

An analysis of the dependence of the average pollutant immission on the average pollutant emission rate has shown that the modelling of pollutant emissions may be

used for the purposes of assessment of environmental risk [4, 10]. For this reason, even actual modelling of a pollutant immission may be classified in the category of modelling of pollutant emissions, because in all the other cases, the modelling of pollutant emissions is indispensable for the assessment of a pollutant immission.

The models of immission of specific size fractions of the particulate matter coming from automotive sources are built with an assumption of functional similarity ("behaviouristic models") [4, 9–14].

For the particulate matter PM10, the relationship between the immission of particulate matter PM10 and the immissions of nitrogen oxides NO_x (i.e. nitrogen oxide NO and nitrogen dioxide NO_2 calculated in terms of nitrogen dioxide equivalent) and carbon monoxide CO is used [4, 10–14]. Such an assumption arises from the postulates that the particulate matter emission related to motor traffic increases with the emissions of nitrogen oxides and carbon monoxide and that the average pollutant immission is an increasing function of the average pollutant emission rate. Actually, a factor conducive to higher particulate matter emission related to motor traffic is the high vehicle speed and, in consequence, high loads of IC engines, which result in high emissions of nitrogen oxides and carbon monoxide [4]. Thus, such a postulate is physically justified. It is known from the modelling of pollutant dissemination, in turn, that an increase in the average pollutant emission rate results in a growth in the average immission of the pollutant involved [4].

In general, the model of immission of the particulate matter PM10 coming from road transport has been assumed to have the form of a function [4, 10, 11, 13, 14]:

$$I_{\text{PM10}} = f_{\text{PM10-NO}_x\text{-CO}}(I_{\text{NO}_x}, I_{\text{CO}}) \quad (1)$$

where:

I_{PM10} – immission of particulate matter PM10;
 I_{NO_x} – immission of nitrogen oxides;
 I_{CO} – immission of carbon monoxide.

This function meets conditions (2) within the range of the immission values that are consistent with the values obtained from the empirical survey and used for model identification purposes [4, 11, 13, 14].

$$\frac{\partial I_{\text{PM10}}}{\partial I_{\text{NO}_x}} > 0; \quad \frac{\partial I_{\text{PM10}}}{\partial I_{\text{CO}}} > 0 \quad (2)$$

The particulate matter immission models, built in accordance with the criterion of functional similarity, are predominantly presented in the literature in the linear form. Consistently, the immission of particulate matter PM10 is modelled as a linear function of the immission of nitrogen oxides in the form as follows [4, 10, 11, 13, 14]:

$$I_{\text{PM10}} = a_{11} + a_{21} \cdot I_{\text{NO}_x} \quad (3)$$

A linear dependence of the immission of particulate matter PM10 on the immission of nitrogen dioxide is postulated as well [4, 10, 11, 13, 14].

Again, the immission of particulate matter PM10 is modelled in the following form as a linear function of the immission of carbon monoxide [4, 10, 11, 13, 14]:

$$I_{PM10} = a_{12} + a_{32} \cdot I_{CO} \quad (4)$$

Among many possible forms of the functions that meet conditions (2), the structure of the function $F_{PM10}^{-NO_x-CO}$ may also be assumed as a polynomial where the immission of nitrogen oxides and the immission of carbon monoxide would be the arguments [4, 10, 11, 13, 14].

– Polynomial of the first degree:

$$I_{PM10} = a_{13} + a_{13} \cdot I_{NO_x} + a_{33} \cdot I_{CO} \quad (5)$$

– Polynomial of the second degree:

$$I_{PM10} = a_{14} + a_{14} \cdot I_{NO_x} + a_{24} \cdot I_{CO} + a_{34} \cdot I_{NO_x}^2 + a_{44} \cdot I_{CO}^2 + a_{54} \cdot I_{NO_x} \cdot I_{CO} \quad (6)$$

The behaviouristic models of the immission of particulate matter PM10, having been directly derived from empirical data, are very effective and offer very good conformity with the phenomena modelled, but only in the conditions of identification of such models [4, 10, 11, 13, 14]. However, a serious shortcoming of such models is the fact that they are not universal in the situations where the structure and intensity of vehicle traffic, pollutant dissipation conditions and background of the pollutants coming from non-automotive sources are defined. This means that the coefficients of these models may be different in the case of e.g. different routes, road junctions, and modelling periods.

In spite of these difficulties, it seems reasonable to adopt a statistical approach to the issue of identification of behaviouristic models and, in consequence, to develop a standard for sets of behaviouristic models of the immission of specific size fractions of particulate matter for representative conditions of pollutant emission and dissemination.

For the identification of coefficients of the particulate matter PM10 immission models that are to be used for inventorying pollutant emissions, results of empirical surveys covering a period of at least one year should preferably be taken into account [4, 10, 11, 13, 14]. The adoption of a one-year period makes the model independent of the weather conditions that are assumed to change in annual cycles. The model coefficients that have been identified in result of surveys carried out for at least one year will thus be averaged in relation to varying weather conditions.

A different situation takes place if the location of the areas of modelling is the matter of concern. In such a case, the situations that can occur must be reasonably categorized according to the criteria that describe e.g. the structure and intensity of vehicle traffic, pollutant dissipation conditions, and background of the pollutants coming from sources other than motor transport.

The environmental risks posed by particulate matter PM2.5 and PM1 are also investigated with employing models built in accordance with the functional similarity criterion [4, 9, 12, 14]. Definitions of particulate matter PM10, PM2.5, and PM1 are used for this purpose.

According to the definition of particulate matter fractions, the set of particulate matter PM_{2.5} is a subset of particulate matter PM₁₀. Based on results of empirical surveys on the immission of particulate matter PM_{2.5} and PM₁₀, a linear dependence of the immission of particulate matter PM_{2.5} on the immission of particulate matter PM₁₀ may be postulated [4, 9, 12, 14]:

$$I_{PM2.5} = k_{PM2.5-10} \cdot I_{PM10} \quad (7)$$

where:

$k_{PM2.5-10}$ – coefficient of the particulate matter PM_{2.5} immission model; $k_{PM2.5-10} \in \langle 0; 1 \rangle$.

Like in the case of modelling the particulate matter PM_{2.5} immission, particulate matter PM₁ may be treated as a subset of particulate matter PM₁₀ and of particulate matter PM_{2.5}. Consistently, the immission of particulate matter PM₁ may be modelled as linearly dependent on the immission of particulate matter PM₁₀ [4, 12, 14]:

$$I_{PM1} = k_{PM1-10} \cdot I_{PM10} \quad (8)$$

where:

k_{PM1-10} – coefficient of the particulate matter PM₁ immission model; $k_{PM1-10} \in \langle 0; 1 \rangle$,

and as linearly dependent on the immission of particulate matter PM_{2.5} [4, 12, 14]:

$$I_{PM1} = k_{PM1-2.5} \cdot I_{PM2.5} \quad (9)$$

where:

$k_{PM1-2.5}$ – coefficient of the particulate matter PM₁ immission model; $k_{PM1-2.5} \in \langle 0; 1 \rangle$.

The identification of the particulate matter PM_{2.5} and PM₁ immission models consists in determining the coefficients $k_{PM2.5-10}$, k_{PM1-10} , and $k_{PM1-2.5}$, based on results of empirical surveys of the immissions of particulate matter fractions PM₁₀, PM_{2.5}, and PM₁. In general, the identification results depend on the pollutant emission and dissipation conditions and on the period during which the measurements were carried out.

The particulate matter is emitted from many different sources. Therefore, the coefficients of the particulate matter PM_{2.5} and PM₁ immission models may be expected to depend on the quantities that characterize the pollutant emission sources and dissemination conditions. In particular, significant differences may be expected to occur between the values of these coefficients for areas with different land relief and development and for areas with diversified intensity of industrial activities and motor transport. Therefore, a statistical approach to the identification of the particulate matter PM_{2.5} and PM₁ immission models is advisable.

The particulate matter PM₁₀ and PM_{2.5} immission models were identified as an example. So far, the immission of particulate matter PM₁ has not been regularly surveyed within the activities of the State Environmental Monitoring. Results of surveys carried out at two measuring stations of the air quality monitoring network in the Małopolska Region in 2012 were used for the model identification purposes [33]. One of them, situated in Aleja Krasieńskiego in Cracow, is an on-road monitoring station installed in a street canyon.

The other one is placed in ulica Bulwarowa in Nowa Huta, close to a housing estate; this location was chosen to investigate the impact of industrial facilities on ambient air quality.

Single measurement results were missing in the set of data available; therefore, the missing data points were estimated by interpolation. Moreover, the pollutant immission vs. time curves were smoothed, i.e. high-frequency noise was removed, with the use of a second-order non-recursive filter.

Figs. 1–4 show time histories of the immissions of particulate matter PM10, nitrogen oxides, and carbon monoxide as recorded at individual air quality monitoring stations on specific days [d].

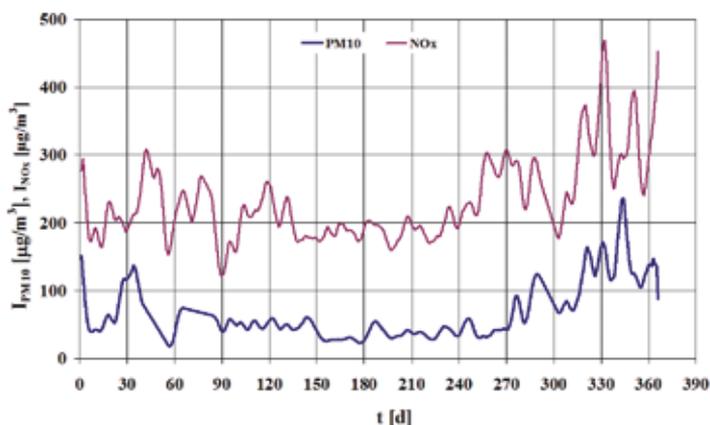


Fig. 1. Immissions of particulate matter PM10 and nitrogen oxides at the air quality monitoring station in Al. Krasieńskiego (Cracow)

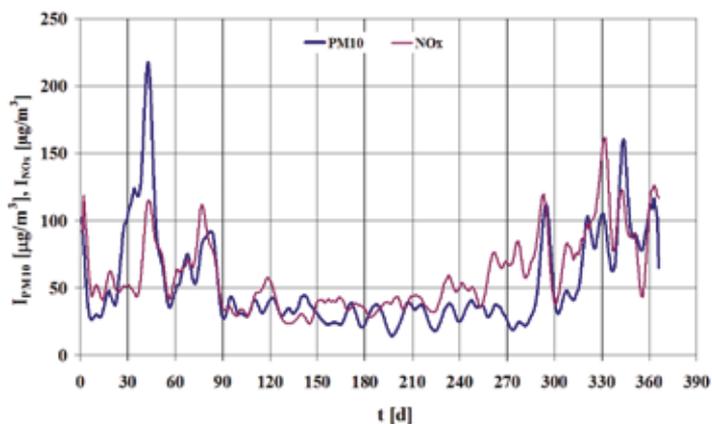


Fig. 2. Immissions of particulate matter PM10 and nitrogen oxides at the air quality monitoring station in Nowa Huta

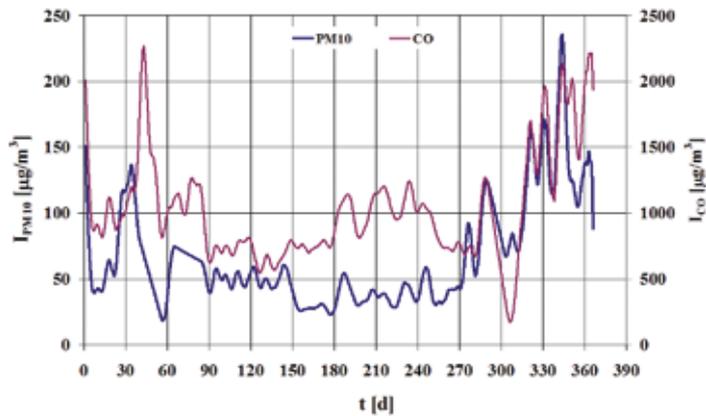


Fig. 3. Immissions of particulate matter PM10 and carbon monoxide at the air quality monitoring station in Al. Krasieńskiego (Cracow)

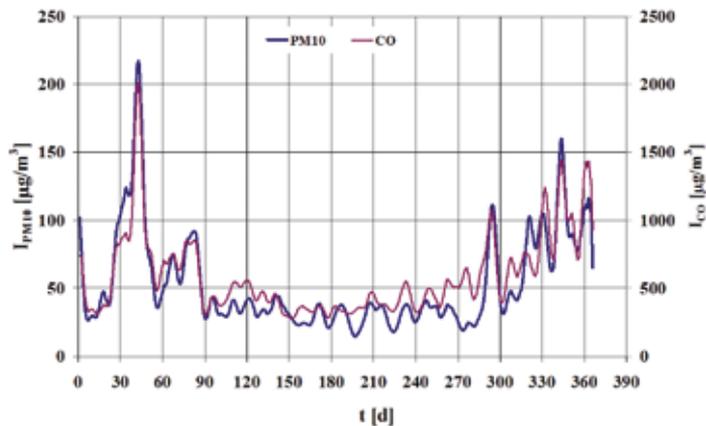


Fig. 4. Immissions of particulate matter PM10 and carbon monoxide at the air quality monitoring station in Nowa Huta

Correlation dependences of the particulate matter immissions on the immissions of nitrogen oxides and carbon monoxide have been presented in Figs. 5–8. The linear functions approximating the dependences of the PM10 particulate matter immission on the immissions of nitrogen oxides and carbon monoxide have also been visualized on the graphs.

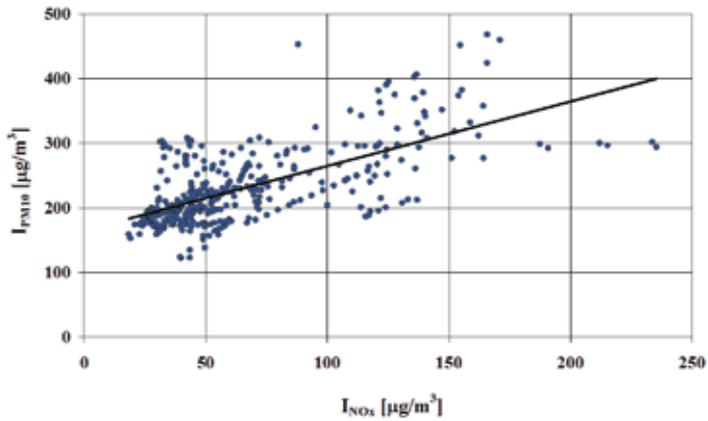


Fig. 5. Correlation dependence of the immission of particulate matter PM10 on the immission of nitrogen oxides at the air quality monitoring station in Al. Krasieńskiego (Cracow)

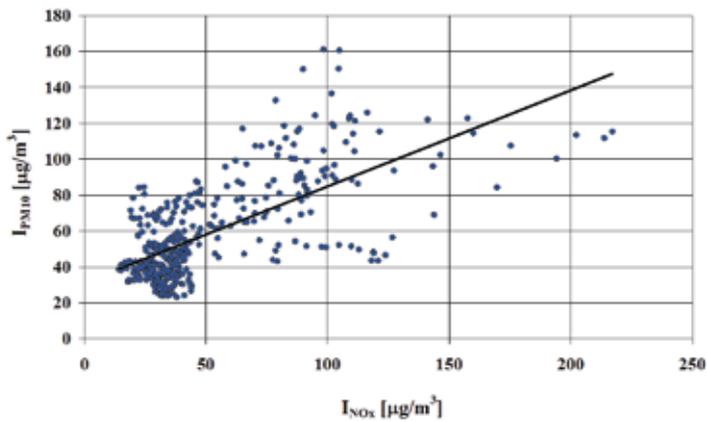


Fig. 6. Correlation dependence of the immission of particulate matter PM10 on the immission of nitrogen oxides at the air quality monitoring station in Nowa Huta

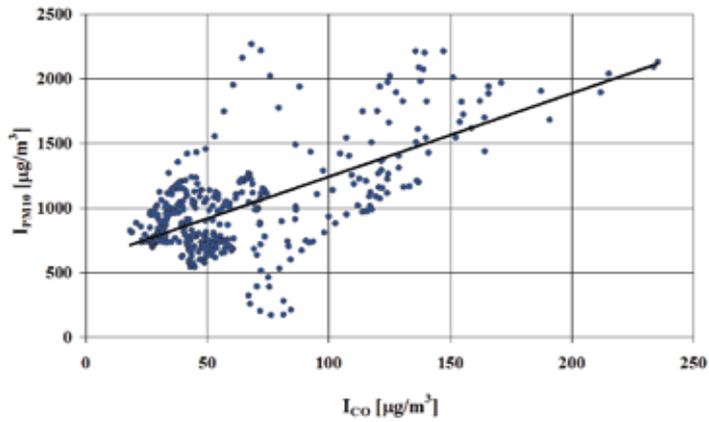


Fig. 7. Correlation dependence of the immission of particulate matter PM10 on the immission of carbon monoxide at the air quality monitoring station in Al. Krasieńskiego (Cracow)

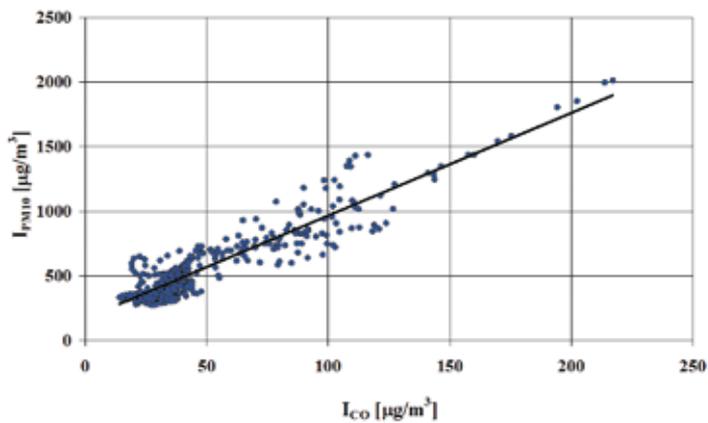


Fig. 8. Correlation dependence of the immission of particulate matter PM10 on the immission of carbon monoxide at the air quality monitoring station in Nowa Huta

The hypothesis on a correlation between the data sets under investigation was verified with the use of a statistical test based on a theory developed by Pearson [27]. The Pearson's coefficient of linear correlation between the data sets under investigation has been presented in Fig. 9.

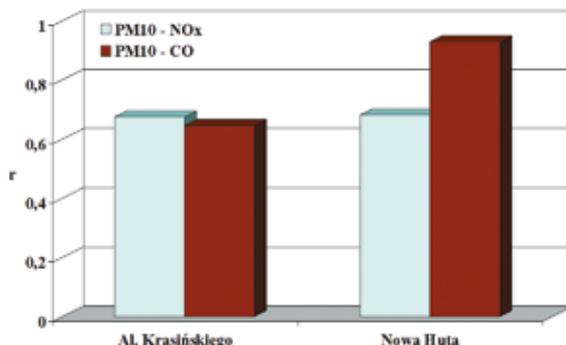


Fig. 9. Pearson's coefficient of linear correlation between the sets of immissions of particulate matter PM10, nitrogen oxides, and carbon monoxide

Evaluation of the Pearson's coefficient of linear correlation did not give unequivocal grounds for postulating any differences in the quality of the correlation between the data sets under investigation. In spite of significant differences in values of the linear correlation coefficient, the probability that the hypothesis on absence of a correlation will not be rejected was found to be below 0.01 for all the tested combinations of the data sets.

The values of the coefficients of models (3) and (4) were determined with the use of the least squares method [23]. The values of the coefficients of models (3) and (4) have been given in the Table 1 below.

Table 1. Coefficients of models (3) and (4), determined for the air quality monitoring stations in Al. Krasieńskiego (Cracow) and in Nowa Huta

Coefficient	Al. Krasieńskiego (Cracow)		Nowa Huta	
	Model (3)	Model (4)	Model (3)	Model (4)
a_{11}, a_{12}	0.998	6.46	0.535	7.95
a_{21}, a_{32}	165.0	596.0	31.32	170.4

The differences in the model coefficient values determined for the two air quality monitoring stations under consideration confirm high susceptibility of these coefficients to the conditions of pollutant emission from motor transport and the other sources and to the conditions of dissemination of these pollutants.

Time histories of the immissions of particulate matter PM10 and PM2.5 at the air quality monitoring stations have been shown in Figs. 10 and 11.

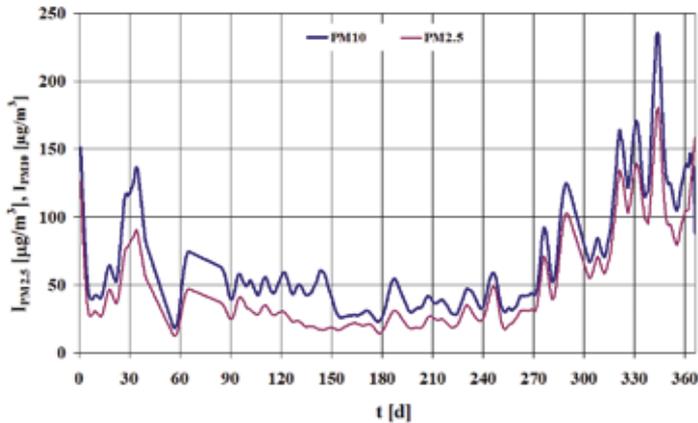


Fig. 10. Immissions of particulate matter PM2.5 and PM10 at the air quality monitoring station in Al. Krasńskiego (Cracow)

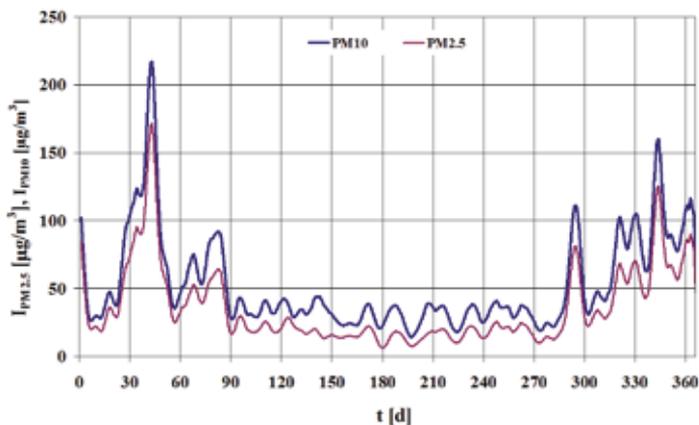


Fig. 11. Immissions of particulate matter PM2.5 and PM10 at the air quality monitoring station in Nowa Huta

Correlation relationships between the immissions of particulate matter PM10 and PM2.5 at the air quality monitoring stations under consideration, with graphs representing the linear functions approximating these relationships, have been presented in Figs. 12 and 13.

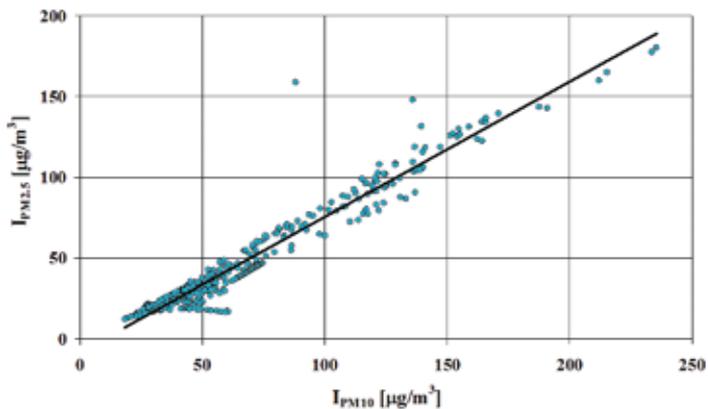


Fig. 12. Correlation relationship between the immissions of particulate matter PM2.5 and PM10 at the air quality monitoring station in Al. Krasińskiego (Cracow)

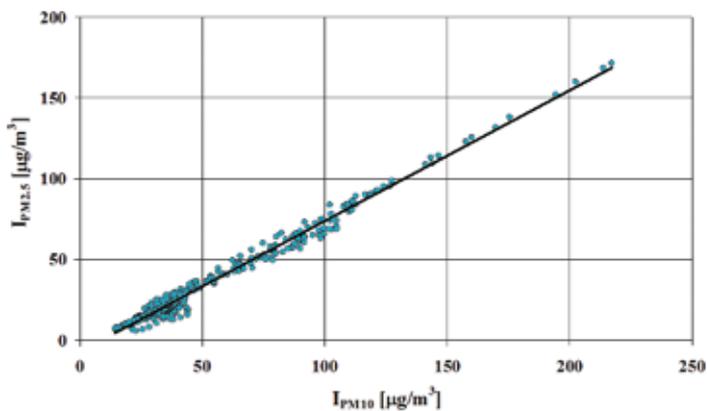


Fig. 13. Correlation relationship between the immissions of particulate matter PM2.5 and PM10 at the air quality monitoring station in Nowa Huta

The coefficient of correlation between the analysed sets of particulate matter PM10 and PM2.5 immission data has been presented in Fig. 14.

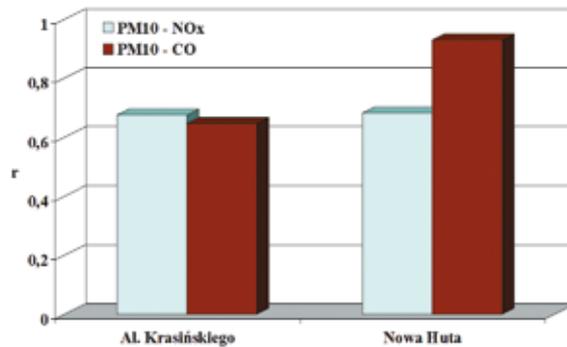


Fig. 14. Pearson's coefficient of linear correlation between the sets of immissions of particulate matter PM10 and PM2.5

For both the air quality monitoring stations, the values of the Pearson's coefficient of linear correlation between the sets of immissions of particulate matter PM10 and PM2.5 are close to 1. The probability that the hypothesis on absence of a correlation will not be rejected was found in both cases to be below 0.01. The correlation test results provide grounds for postulating strong linear dependence of the immission of particulate matter PM2.5 on the immission of particulate matter PM10.

Time histories of the coefficient of the particulate matter PM2.5 immission model, determined for the two air quality monitoring stations, with the annual average values of this coefficient have been shown in Figs. 15 and 16.

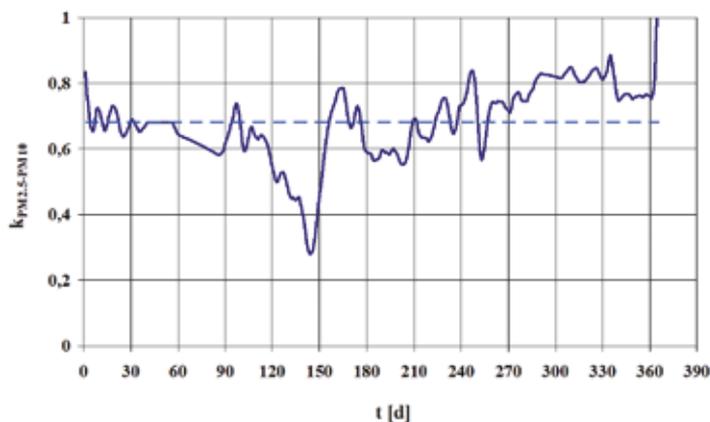


Fig. 15. Time history and annual average value of the coefficient of the particulate matter PM2.5 immission model for the air quality monitoring station in Al. Krasińskiego (Cracow)

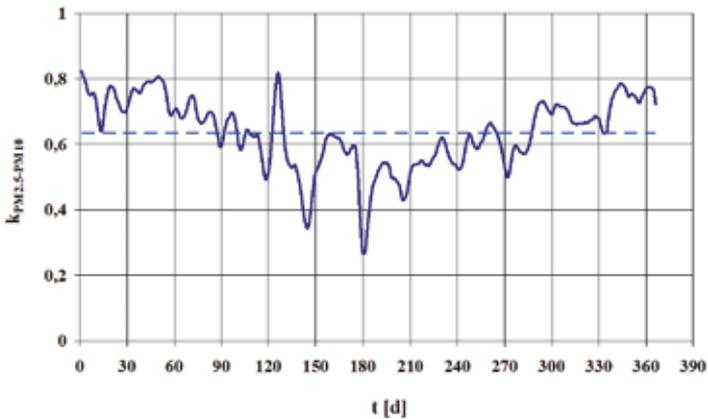


Fig. 16. Time history and annual average value of the coefficient of the particulate matter PM2.5 immission model for the air quality monitoring station in Nowa Huta

For both the air quality monitoring stations, the values of the coefficient of the particulate matter PM2.5 immission model are close to each other and they exceed 0.6. In the summer months, these values are lower than those recorded in the winter season. This probably can be explained by the fact that an important reason for the emission of very fine particles in winter is the combustion of solid fuels for heating purposes.

4. Recapitulation

Dusts pose serious danger to the human health and environment. This is confirmed by numerous research works carried out by independent institutions all over the world. It has been unequivocally ascertained that particularly harmful health impact is exerted by respirable dusts, which deeply penetrate into the respiratory system. In connection with the risks of this kind, increasingly stringent regulations are introduced to protect the human health and environment from dusts. Originally, the regulations were only applicable to particulate matter fraction PM10; at present, they also cover the PM2.5 fraction. The exceedances of the immissions of specific particulate matter size fractions are the most frequent situations of risk to the ambient air quality, especially in the central parts of large urban agglomerations. Therefore, a necessity exists to monitor the ambient air quality in terms of immissions of specific particulate matter size fractions.

Empirical measurements are only possible within a limited scope, usually on the premises of air quality monitoring stations. In other areas, the environmental risks caused by dusts may only be assessed by modelling pollutant emissions and dissemination. Pollutant emissions may be modelled for both natural and civilization processes. The emission of pollutants from stationary sources usually can be identified. The modelling of pollutant

emissions from mobile sources, especially from motor vehicles, is a much more difficult task while this emission is a basic component of the total pollutant emission in the central parts of large urban agglomerations, i.e. in the areas of particularly high risk to large numbers of people. The emission of pollutants from motor vehicles is modelled with the use of modelling the motor vehicle traffic conditions and the characteristics of pollutant emissions from motor vehicles. The assessment of ambient air quality is based on pollutant immission values. The pollutant immissions are determined from the rates (intensity) of emission of individual pollutants, with the use of pollutant dissemination models [4].

From the point of view of the risks posed by dusts, two methods are used to model the emissions and immissions of specific size fractions of the particulate matter coming from motor vehicle traffic. For the PM₁₀ particulate matter fraction, emission models based on structural (morphological) similarity are applied [4, 10, 33]. To model the immission of particulate matter PM₁₀, models built in accordance with the functional similarity (behaviouristic models) are employed as well [4, 8, 10–14]. The functional similarity is also taken into account to model the immissions of particulate matter PM_{2.5} and PM₁ [9, 12, 14, 33].

The modelling of the immission of particulate matter PM₁₀ is based on the postulated dependence of the emission of particulate matter from motor transport sources on the emissions of nitrogen oxides and carbon monoxide [4, 8, 10–14]. Consistently with taking into account the increasing dependence of the average pollutant immission on the average pollutant emission rate, an assumption is made that the immission of particulate matter PM₁₀ depends on the immissions of nitrogen oxides and carbon monoxide. The results of empirical surveys of the immissions of particulate matter PM₁₀, nitrogen oxides, and carbon monoxides confirm that a considerable correlation exists between the analysed sets of data on the immission of these pollutants. In the literature, linear models of the immission of particulate matter PM₁₀ vs. the immissions of nitrogen oxides and carbon monoxide can be most frequently met. These models may also be generalized to the form of two-parameter non-linear models. The research to date has not revealed a significant difference in effectiveness between one-parameter linear models and non-linear models, whether of the one-parameter or two-parameter type [4, 10, 13].

The effectiveness of models of the immission of particulate matter PM₁₀ is chiefly limited by difficulties in the identification of such models, as the model coefficient values that can be met in the literature range within very wide limits and are very difficult to be associated with physical interpretation of motor traffic [4, 10].

The modelling of the immission of particulate matter PM_{2.5} is based on the postulated linear dependence of the immission of particulate matter PM_{2.5} on the immission of particulate matter PM₁₀; similarly, the modelling of the immission of particulate matter PM₁ is based on the postulated linear dependence of the immission of particulate matter PM₁ on the immission of particulate matter PM_{2.5} and on the immission of particulate matter PM₁₀ [9, 12, 14, 33]. The results of empirical surveys confirm that a considerable correlation exists between the immissions of these pollutants. These surveys also unequivocally indicate that the predominating source of the very fine dusts, i.e. particulate matter PM_{2.5} and PM₁, is not the motor transport but the power industry, ranging from large heat and

power plants to dispersed municipal and household sources, which is related, above all, to the use of solid fuels for heating purposes [4, 12, 14].

The effectiveness of the pollutant immission models directly based on results of empirical surveys carried out in the areas of motor traffic is very good, but only in the conditions of measurements of pollutant immissions. This means that the coefficients of such models are susceptible both to the pollutant emission conditions, which depend on the motor traffic, and to the pollutant dissemination conditions. This finding has been confirmed by the investigation results presented herein and by many other results published in the literature quoted [4, 8–14, 33]. In spite of such a limitation, it seems reasonable to believe that the modelling of the immission of specific particulate matter size fractions may be considered an effective method to be employed when investigating the risk posed by dusts to the human health and environment.

Acknowledgements

The publication has been prepared with using results of a research and development project No. NR10-0050-10/2010 entitled "Development of systems to reduce dust emission from disc brake and drum brake mechanisms of automotive vehicles," financed from the funds of the National Centre for Research and Development.

References

- [1] AGRICOLA G.: *De re metalica*. <http://www.farlang.com/gemstones/agricola-metallica/>.
- [2] CANAGARATNA M.: *Chase studies of particulate emissions from in-use New York City vehicles*. *Aerosol Science and Technology* 2004, 38(6), 555–573.
- [3] CAPES C. E.: *Particle size enlargement*. Elsevier Sci. Publ. Co., Amsterdam 1980.
- [4] CHŁOPEK Z. et al.: *Modelowanie emisji cząstek stałych PM10 ze źródeł motoryzacyjnych do celów oceny oddziaływania transportu drogowego na środowisko (Modelling of the emission of particulate matter PM10 from automotive sources for the purposes of environmental impact assessment of road transport)*. Report of research project No. N N509 083637 sponsored by the Ministry of Science and Higher Education, Warszawa 2012 (not published, in Polish).
- [5] CHŁOPEK Z., JAKUBOWSKI A., KIERACIŃSKA A.: *Examination of a laboratory system to reduce dust emission from braking systems of automotive vehicles*. *The Archives of Automotive Engineering – Archiwum Motoryzacji* 3/2011, 5–17, 103–115.
- [6] CHŁOPEK Z., JAKUBOWSKI A.: *A study of the particulate matter emission from the braking systems of motor vehicles*. *Eksplatacja i Niezawodność – Maintenance and Reliability* No. 4 (44)/2009, 45–52.
- [7] CHŁOPEK Z., JAKUBOWSKI A.: *The examination of the reduction of particulate matter emission from motor vehicle braking systems*. *Eksplatacja i Niezawodność – Maintenance and Reliability* No. 4(48)/2010, 29–36.
- [8] CHŁOPEK Z., PIASECZNY L.: *The modelling of the particulate matter PM10 emission in port cities*. *Maritime Transport. Technological Innovations & Research*. Universitat Politècnica de Catalunya, BARCELONATECH, Barcelona 2012, 661–670.
- [9] CHŁOPEK Z., SKIBIŃSKI F.: *Wprowadzenie do tematyki emisji cząstek stałych PM2.5 z transportu samochodowego (Introduction to the subject matter of the emission of particulate matter PM2.5 from motor transport)*. *Transport Samochodowy* 3/2010, 73–87 (in Polish).

- [10] CHŁOPEK Z., ŻEGOTA M.: *The emission of particulate matter PM10 from vehicles*. Eksploatacja i Niezawodność – Maintenance and Reliability No. 1 (21)/2004, 3–13.
- [11] CHŁOPEK Z.: *Examination of a particulate matter PM10 immission model in the environment around road transport routes*. The Archives of Automotive Engineering – Archiwum Motoryzacji 1/2012, 23–38, 121–136.
- [12] CHŁOPEK Z.: *Identyfikacja modeli emisji cząstek stałych PM2.5 i PM1 (The identification of particulate matter PM2.5 and PM1 immission models)*. Zeszyty Naukowe Instytutu Pojazdów, The Warsaw University of Technology 1(87)/2012, 26–31 (in Polish).
- [13] CHŁOPEK Z.: *Modele behawiorystyczne emisji cząstek stałych PM10 ze źródeł transportu drogowego (Behavioural models of the PM 10 particulates emission from the road transport sources)*. Zeszyty Naukowe Instytutu Pojazdów, The Warsaw University of Technology 1(82)/2011, 111–118 (in Polish).
- [14] CHŁOPEK Z.: *Testing of hazards to the environment caused by particulate matter during use of vehicles*. Eksploatacja i Niezawodność – Maintenance and Reliability, 2/2012, 160–170.
- [15] DUBIELLA–JACKOWSKA A. et al.: *Skład zanieczyszczeń wprowadzanych do środowiska wzdłuż dróg o różnym natężeniu ruchu pojazdów mechanicznych (Composition of pollutants in the water samples collected along the highways with different traffic intensity)*. Ecological Chemistry and Engineering vol. 14, No. S3, 2007 (in Polish).
- [16] EN 481:1993. *Workplace atmospheres. Size fraction definitions for measurement of airborne particles*.
- [17] FORSBERG B. et al.: *Comparative health impact assessment of local and regional particulate air pollutants in Scandinavia*. Journal of the Human Environment 34(1)2005, 11–19.
- [18] ISO 7708:1995. *Air quality. Particle size fraction definitions for health-related sampling*.
- [19] JÓŹWIAK S., MIZAK K., ZASADA D., JÓŹWIK P.: *Badania próbek pyłów z tarczowych i bębnowych układów hamulcowych pojazdów samochodowych (The study of samples of dust from disc and drum brake systems of motor vehicles)*. Report No. 056–58/LBM/2012, Military University of Technology, Warszawa 2012 (not published, in Polish).
- [20] JÓŹWIAK S., MIZAK K., ZASADA D.: *Badania dwóch próbek materiału ciernego: z tarczowych i bębnowych układów hamulcowych (The study of two samples of brake friction material from disc and drum brake systems of motor vehicles)*. Report No. 056–60/LBM/2012, Military University of Technology, Warszawa 2012 (not published, in Polish).
- [21] *Leksykon ekoinżynierii (Lexicon of eco-engineering)*. Edited by Gabriel Borowski, Polskie Towarzystwo Inżynierii Ekologicznej (PTIE – Polish Society of Ecological Engineering), Warszawa 2010 (in Polish).
- [22] MACNEE W., DONALDSON K.: *Exacerbations of COPD – environmental mechanism*. Chest 2000, 117, 390–397.
- [23] MAŃCZAK K.: *Metody identyfikacji wielowymiarowych obiektów sterowania (Methods of identification of multidimensional objects of control)*. Warszawa, Wydawnictwo Naukowo–Techniczne, 1971.
- [24] MAYER A. et al.: *Nanoparticle–emissions of EURO 4 and EURO 5 HDV compared to EURO 3 with and without DPF*. SAE Emissions Measurement and Testing. 2007. 335–43.
- [25] NAWROT T. S. et al.: *Public health importance of triggers of myocardial infarction: a comparative risk assessment*. The Lancet 2011, vol. 377, No. 9767, 732–740.
- [26] PAŃCZYK M., GIRO L.: *Wykonanie zdjęć skaningowych (BSE i SE) i oznaczenie składu chemicznego pyłów w mikroobszarze z wykorzystaniem skaningowego mikroskopu elektronowego LEO sprzężonego z EDX (Making scanning photography (BSE and SE) and estimation of the chemical composition of dust in micro area using scanning electron microscopy coupled with EDX)*. Report No. 45.2900.1224.49.2, Państwowy Instytut Geologiczny (PGI – Polish Geological Institute), Warszawa 2012 (not published, in Polish).
- [27] PEARSON K.: *On the theory of contingency and its relation to association and normal correlation*. Drapers' Company Research Memoirs, Biometric 1904, Ser. I.
- [28] PIELECHA J.: *Identyfikacja parametrów cząstek stałych w silnikach spalinowych (Identification of parameters of particulate matter in internal combustion engines)*. Wydawnictwo Politechniki Poznańskiej (Publishing House of the Poznań University of Technology), Series "Dissertations" No. 67, Poznań 2012 (in Polish).
- [29] Pietsch W.: *An interdisciplinary approach to size enlargement by agglomeration*. Powder Technology, 130/2003, 8–13.
- [30] RADWAG: *Oznaczenie pyłu całkowitego i respirabilnego metodą filtracyjno – wagową na stanowiskach pracy (Determination of total and respirable dust at work stations by a filtration–mass method)*. http://www.radwag.pl/e-sklep/pliki/artykuly/oznaczanie_pyłu.pdf (in Polish).

- [31] *Rozporządzenie Ministra Pracy i Polityki Społecznej z dnia 29 listopada 2002 r. w sprawie najwyższych dopuszczalnych stężeń i natężeń czynników szkodliwych dla zdrowia w środowisku pracy (Regulation of the Minister of Labour and Social Policy of November 29, 2002 on the maximum permissible concentrations and intensities of harmful factors in the work environment)*. Dz. U. 2002 No. 217, item 1833 (in Polish).
- [32] SIEMIŃSKI M.: *Środowiskowe zagrożenie zdrowia (Environmental health risk)*. PWN, Warszawa 2001 (in Polish).
- [33] SUCHOCKA K.: *Modelowanie imisji frakcji wymiarowych cząstek stałych ze względu na oddziaływanie motoryzacji na środowisko (Modelling of the immission of specific particulate matter size fractions from the point of view of the environmental impact of motorization)*. Master's graduation work, Warsaw University of Technology, Faculty of Environmental Engineering, Warszawa 2013 (not published, in Polish).