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SEASONAL VARIABILITY OF CONCENTRATIONS OF TOTAL SUSPENDED PARTICLES (TSP) AS WELL AS PM10, PM2.5 AND PM1 MODES IN ZABRZE, POLAND

KRZYSZTOF KLEJNOWSKI, ANDRZEJ KRASA, WIOLETTA ROGULA

Institute of Environmental Engineering of the Polish Academy of Sciences
ul. M. Skłodowskiej-Curie 34, 41-819 Zabrze, Poland

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SEZONOWA ZMIENNOŚĆ STĘŻEŃ CAŁKOWITEGO PYŁU ZAWIESZONEGO (TSP) ORAZ PM10, PM2.5 I PM1 W ZABRZU

W Instytucie Podstaw Inżynierii Środowiska PAN od stycznia 2006 r. prowadzone są pomiary rozkładu ziarnowego pyłu zawieszonego, w tym mierzone są stężenia frakcji < 1 μm (PM1) z użyciem impaktora PM10 firmy Dekati. W pracy przedstawiono otrzymane w 2006 r. wyniki – skład frakcyjny całkowitego pyłu zawieszonego z uwzględnieniem częstości występowania poszczególnych przedziałów stężeń PM1 w sezonie letnim i zimowym w punkcie „tła miejskiego" w Aglomeracji Górnośląskiej w Zabrzu. Przedstawiono obliczone średnie miesięczne i sezonowe TSP oraz stężeń frakcji PM10, PM2,5 i PM1. Określono udział frakcji grubej, drobnej i PM1 w pyle zawieszonym.

Summary

From January 2006 on, a continuous experiment consisting in determinations of granulometric composition of the airborne dust has been performed in the Institute of Environmental Engineering of PAS, Zabrze. The investigations include measurements of concentrations of PM1 – the suspended in ambient air particles of the aerodynamic diameter not grater than 1 μm – by using the PM10 Dekati impactor. The results of twelve month measurements (January – December 2006) are presented in the paper: the granulometric composition of total suspended dust (PM10, PM2.5, PM1) in winter and summer at an urban background site in the Silesian Agglomeration. Monthly and seasonal average concentrations of PM10, PM2.5 and PM1 were computed.

INTRODUCTION

Solid ambient particulates have been investigated for many years. The scope of research widens systematically following development of new measurement methods and analytical techniques that allow for more precise measurements. Along with possibility of collecting and developing greater sets of data, their higher quality enables to include human health problems and epidemiological aspects of exposure of inhabitants of urban areas within the interest of investigators. Suspended dust – in its fractions PM10, PM2.5, PM1 defined by their physicochemical properties [25] – as well as chemicals adsorbed

onto surface of its particles has always been considered harmful to humans. The researchers' last decade interest in PM10 yielded many results that led to establishing proper air quality standards [2, 10, 29]. Epidemiological studies and other investigations concerning influence of suspended dust firmly confirm particularly adverse effects of the finest dust fraction. The finest particles penetrate into the deepest regions of lungs to where they transport toxic substances [5, 13, 23, 24]. In some circumstances, some carcinogens occur exclusively on particles having aerodynamic diameters less than 2.0 μm [5, 11]. The finest dust particles affect the global climate changes and threaten human health, they change cloud capability of absorbing or reflecting solar light affecting in this way natural energetic balance of the Earth [12].

These observations justify common interest in the solid particles of aerodynamic diameter between 0 and 2.5 μm suspended in the atmospheric air. The aerodynamic diameter equal to 1 μm is the lowest upper bound of the diameters of suspended particles possible to be observed for long periods of time, in great flows of air and by using inertial methods [16, 26]. An important argument in support for the interest in PM1 may also be growing emission from car engines – as well spark as compression ignition ones [9, 21].

In the Air Protection Department of the Institute of Environmental Engineering, during whole 2006 year, concentrations of PM1, PM2.5 and PM10, the fractions of total suspended dust (TSP), were measured with use of the PM10 Dekati cascade impactor. The paper presents results of these investigations reflecting seasonal variability of granularometric composition of TSP during whole 2006 year.

LOCATION OF THE MEASURING POINT

Measurements were performed in the period from 17 January to 30 December 2006. The measuring point was located in the premises of the Institute, in the central part of Zabrze. About 500 m north of the point there is the 88 national highways, a quite busy two-lane road. About 300 m east of the point there is a three-lane road, less busy than the

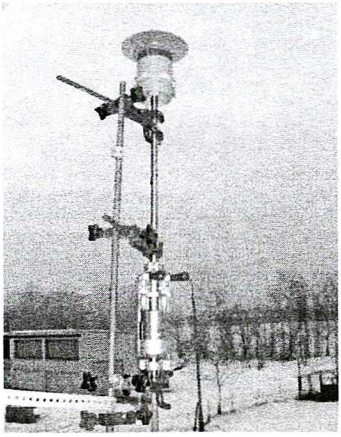

Fig. 1. Measuring equipment in the IEE PAS in Zabrze

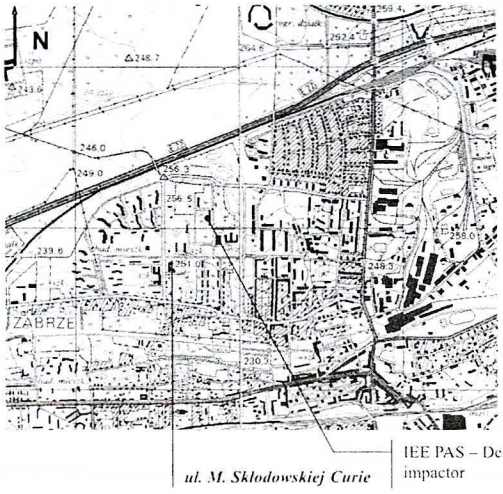

Fig. 2. Location of the measuring point in Zabrze

previous one, and behind it, farther to the east a quarter of high-rise blocks. South-east and south of the measuring point, there are low city houses, built at the beginning of the XX century. About 400 m west of the point there are again very high rise-blocks. The low houses are individually coal-heated; the high blocks are attached to the central heat distribution system.

Also in the Institute premises, the site characteristic of background conditions for air quality monitoring, an automatic air quality monitoring station, belonging to the Regional Inspectorate for Environment Protection [30], is located – the fact confirming the measuring point representatibility of the Zabrze ambient air conditions.

The measuring point location is presented in Figure 2. In Figure 1, the impactor inlet is presented – it is located at about 7 m above the ground to eliminate effects of secondary emission from adjacent area.

METHOD

Dust was collected with use of the three stage Dekati PM-10 impactor which allows for collection of TSP with its simultaneous separation into three fractions: PM10, PM2.5 and PM1. In Figure 3, the impactor working principle is presented.

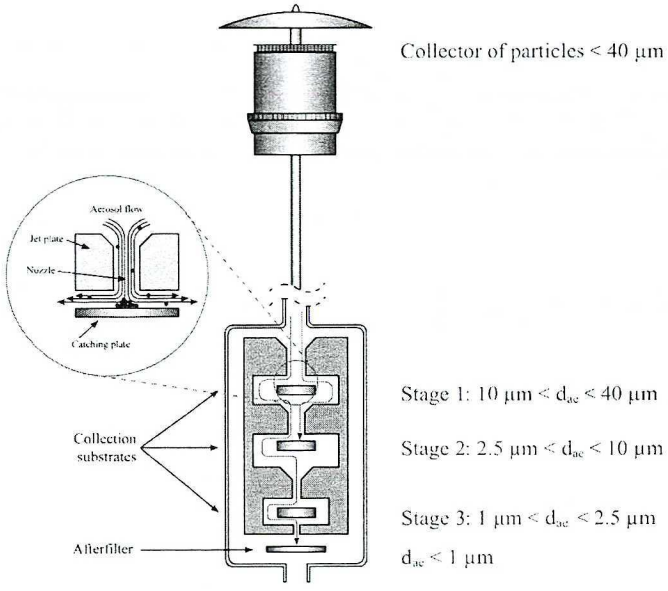

Fig. 3. The Decati PM-10 working principle

The working principle of an impactor is simple. Two parallel plates are applied: upper, a jet plate, with a nozzle bored through it, and lower, a catching plate (Fig. 3, the magnification in the circle). Aerosol passes through the nozzle at high speed. The aerosol makes a sharp turn over the lower plate overflowing it. Less inert particles follow the

stream; more inert ones hit the lower plate where they are caught. A cut diameter of an impactor is the aerodynamic diameter of particles that are caught with 50% efficiency. A real impactor jet plate usually has some number of nozzles and its catching plate is a substrate. Impactor have usually several such pairs of plates arranged in a sequence with decreasing from inlet to outlet cut diameters. Such a set segregates particles by their size while a stream of aerosol passes through the impactor.

The Decati PM-10 impactor has three stages (Fig. 3):

- first stage (the cut diameter 10.0 μm) catches all particles with the aerodynamic diameter greater than 10.0 μm and 50% of particles with 10.0 μm diameter;
- second stage (the cut diameter 2.5 μm) catches all particles with the aerodynamic diameter greater than 2.5 μm, not greater than 10.0 μm (only such particles reach this stage) and 50% of particles with 2.5 μm diameter;
- third stage (the cut diameter 1.0 μm) catches all particles with the aerodynamic diameter greater than 1.0 μm, not greater than 2.5 μm and 50% of particles with 1.0 μm diameter;

and an afterfilter (filter substrate) as the fourth stage:

- it catches all particles that are not caught at the precedent stages, i.e. 50% of particles with aerodynamic diameter equal to 1.0 μm and all smaller ones.

Generally, a single sample collection lasted 48 hours; in periods of heavy air pollution the time of collecting a sample was limited to 24 hours. A sample-taking always started about midnight.

The gravimetric method was used to determine masses of dust samples. At the three first stages, the dust was collected on aluminum or polyurethane 25 mm substrates, at the last one – on 47 mm glass fiber filter. The substrates were conditioned in the weighing room during 48 hours before and after exposure. After conditioning, the substrates were weighed with the use of Mettler Toledo scales. Concentrations of TSP, PM10, PM2.5 and PM1 were determined.

RESULTS – PRESENTATION AND DISCUSSION

There were 122 measurements performed during 2006: 60 in the winter (January – March, October – December) and 62 in summer (April – September).

Basic statistical parameters of the measurement series are presented in Table 1 – the data is statistically developed for each month, winter, summer and whole year separately.

The contributions of PM10, PM2.5, PM1 to TSP (percent, TSP is 100%) in Zabrze are presented in Figure 4 (winter) and Figure 5 (summer).

Table 1. Statistical parameters of the measurement series received by using the Dekati PM10 impactor

January, 8 measurements					
Fraction		TSP	PM10	PM2.5	PM1
Average value	[μg/m³]	152.88	150.80	141.83	108.17
Maximum		254.64	252.04	239.39	204.28
Minimum		71.01	70.27	67.12	39.79
Standard deviation		74.27	73.16	69.02	63.10
February, 16 measurements					
Fraction		TSP	PM10	PM2.5	PM1
Average value	[μg/m³]	79.76	78.40	74.28	57.44
Maximum		194.62	194.28	189.51	160.17
Minimum		28.66	28.19	26.75	16.34
Standard deviation		49.12	48.98	47.26	40.87
March, 11 measurements					
Fraction		TSP	PM10	PM2.5	PM1
Average value	[μg/m³]	83.60	81.62	76.28	55.53
Maximum		133.13	130.67	120.71	86.66
Minimum		23.32	20.80	18.24	14.18
Standard deviation		33.72	33.44	31.50	21.98
April, 10 measurements					
Fraction		TSP	PM10	PM2.5	PM1
Average value	[μg/m³]	72.20	66.21	59.74	47.69
Maximum		111.79	108.46	104.94	94.58
Minimum		38.84	36.31	28.77	23.55
Standard deviation		26.47	27.57	26.56	22.55
May, 13 measurements					
Fraction		TSP	PM10	PM2.5	PM1
Average value	[μg/m³]	42.95	34.95	29.54	24.36
Maximum		91.23	78.92	69.81	58.64
Minimum		12.65	10.45	8.81	7.22
Standard deviation		21.62	18.49	16.45	14.18
June, 9 measurements					
Fraction		TSP	PM10	PM2.5	PM1
Average value	[μg/m³]	35.68	30.45	24.99	19.70
Maximum		62.31	58.13	52.22	47.28
Minimum		10.02	7.98	7.48	6.48
Standard deviation		14.28	13.64	12.05	11.59
July, 12 measurements					
Fraction		TSP	PM10	PM2.5	PM1
Average value	[μg/m³]	44.42	37.21	29.22	22.88
Maximum		72.62	59.23	46.48	35.48
Minimum		16.01	10.60	6.70	3.70
Standard deviation		16.94	15.27	12.84	10.80

August, 9 measurements					
Fraction		TSP	PM10	PM2.5	PM1
Average value	[μg/m³]	22.77	19.47	15.67	11.05
Maximum value		40.32	34.32	27.70	20.79
Minimum value		11.89	10.24	8.08	5.66
Standard deviation		8.03	6.98	5.89	4.63
September, 9 measurements					
Fraction		TSP	PM10	PM2.5	PM1
Average value	[μg/m³]	46.33	39.92	34.00	23,65
Maximum		65.62	57.38	49.78	34,10
Minimum		31.51	21.73	15.78	10,65
Standard deviation		11.34	11.03	10.35	6,60
October, 9 measurements					
Fraction		TSP	PM10	PM2.5	PM1
Average value	[μg/m³]	65.49	59.77	53.12	38.68
Maximum		105.08	97.56	84.06	60.30
Minimum		30.91	27.35	22.10	11.23
Standard deviation		28.22	26.76	25.37	18.97
November, 9 measurements					
Fraction		TSP	PM10	PM2.5	PM1
Average value	[μg/m³]	97.26	93.69	83.12	50.47
Maximum		192.68	185.55	157.06	90.44
Minimum		27.40	26.95	24.16	16.95
Standard deviation		63.65	61.82	53.76	27.27
December, 7 measurements					
Fraction		TSP	PM10	PM2.5	PM1
Average value	[μg/m³]	54.68	52.77	48.13	35.12
Maximum		74.91	71.61	63.71	44.07
Minimum		27.48	26.63	23.83	17.48
Standard deviation		15.63	15.02	13.83	9.29
Winter season, 60 measurements					
Fraction		TSP	PM10	PM2.5	PM1
Average value	[μg/m³]	87.81	85.25	78.88	57.43
Maximum		254.64	252.04	239.39	204.28
Minimum		23.32	20.80	18.24	11.23
Standard deviation		54.35	53.82	50.49	40.06
Summer season, 62 measurements					
Fraction		TSP	PM10	PM2.5	PM1
Average value	[μg/m³]	44.46	38.25	32.32	25.13
Maximum		111.79	108.46	104.94	94.58
Minimum		10.02	7.98	6.70	3.70
Standard deviation		22.52	21.45	20.02	16.74
Year, 122 measurements					
Fraction		TSP	PM10	PM2.5	PM1
Average value	[μg/m³]	65.78	61.36	55.22	41.01
Maximum		254.64	252.04	239.39	204.28
Minimum		10.02	7.98	6.70	3.70
Standard deviation		46.58	46.92	44.62	34.45

Table 2. Relations (contributions, %) between mean concentrations of TSP, PM10, PM2.5 and PM1

Contributions	PM1/TSP	PM1/PM10	PM1/PM2.5	PM2.5/PM10	PM2.5/TSP	PM10/TSP
Year						
Maximum, %	88.86	89.52	92.21	97.54	97.37	99.83
Minimum, %	23.11	34.91	50.80	63.21	41.85	65.70
Average value, %	59.16	65.06	74.39	87.28	79.41	90.44
Summer season						
Maximum, %	84.61	87.21	90.54	96.76	93.87	98.77
Minimum, %	23.11	34.91	55.22	63.21	41.85	65.70
Average value, %	53.82	63.29	76.25	82.71	70.24	84.60
Winter season						
Maximum, %	88.86	89.52	92.21	97.54	97.37	99.83
Minimum, %	30.35	36.41	50.80	71.66	59.74	83.36
Average value, %	64.68	66.89	72.46	92.01	88.88	96.48

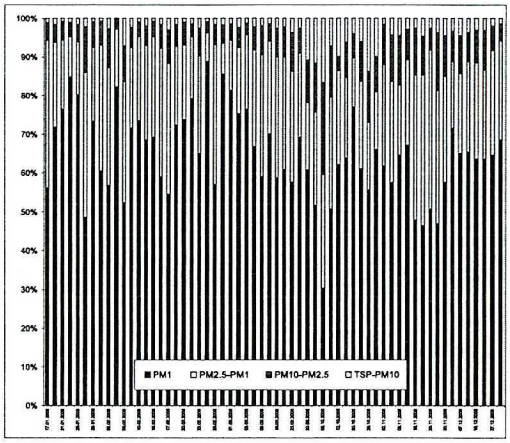

Fig. 4. Contributions of PM10, PM2.5 and PM1 to TSP in winter

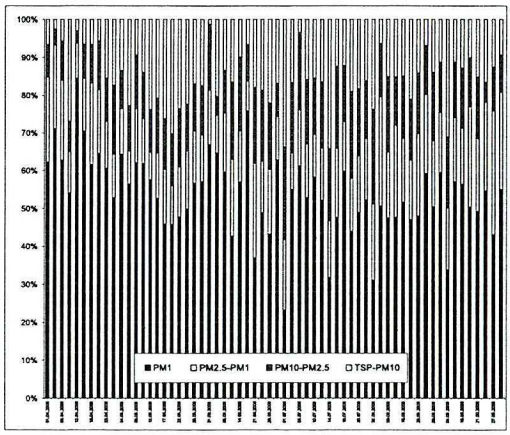

Fig. 5. Contributions of PM10, PM2.5 and PM1 to TSP in summer

The monthly mean concentrations computed for May, June, July, August and September do not exceed the standard equal to 40 μg/m^3. In winter, the PM10 concentrations reach 130–375% of this standard [22] and the mean PM10 winter concentration is 85.25 μg/m^3. The computed mean concentration for 2006 is about 150% of the annual 40 μg/m^3 PM10 standard.

The European Committee proposes 25 μg/m^3 as the permissible annual value for PM2.5 concentration [4]. As it may be seen in Table 1, during the whole year only in June and August this value was not exceeded. The computed annual mean PM2.5 concentration is about 2.2 times this standard.

In Figures 6 and 7, below, histograms of PM2.5 and PM10 are presented.

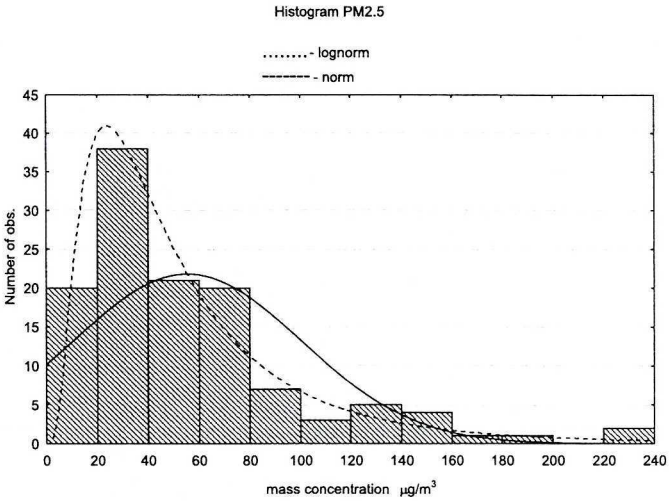

Fig. 6. Frequency distribution of the PM2.5 concentrations in Zabrze, Poland

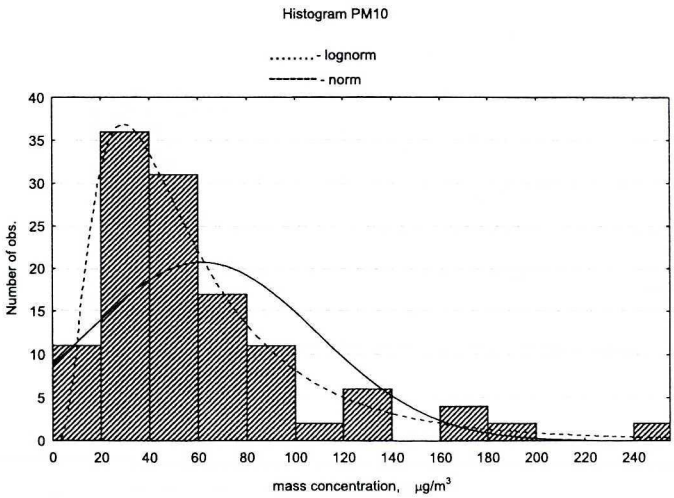

Fig. 7. Frequency distribution of the PM10 concentrations in Zabrze, Poland

As can be seen, a considerable number of PM10 and PM2.5 concentrations in 2006 were high – about 20 results are between 0 and 20 μg/m^3 and almost 40 are between 20 and 40 μg/m^3. It means more than a half of all results greater than 40 μg/m^3. Also more than a half of all PM10 results are between 20 and 60 μg/m^3 and majority of the rest are between 60 and 200 μg/m^3.

These very high concentrations of the two fractions are a result of specific weather and air pollution conditions in January 2006. In this period not only at the Institute but also in many other air monitoring stations over the Silesia Region, values exceeding 24 h standards for PM10 (and SO_2, accidentally also NO_x [18]) were noted. The weather conditions at this time (low temperature, high pressure – downward air movements, very low inversion layer) and heavy air pollution (TSP – Table 1, and also SO_2 and NO_x [20]) allow to consider this situation as a smog episode [19]. Direct observations confirmed very low air transparency and characteristic, very strong, smell of smoke from domestic furnaces. This January 2006 smog episode appeared to be one of the most considerable smog episodes of the recent years in the Silesian Region. The phenomenon was described in [18, 20] in detail.

In Europe, for some last years, various teams of investigators collected data on PM2.5 [3, 17]. In many cases concentrations of PM2.5 and PM10 are very close or even identical [1, 8]. It means that at the sites under investigation amount of coarse particles, i.e. particles with aerodynamic diameter between 2.5 and 10 μm, is small. Similar conclusion may be drawn from the presented investigations. Figures 4 and 5 and Table 2 show that in Zabrze suspended dust consists almost exclusively of fine particles. On average, about 87.28% of PM10 is PM2.5 (maximum 97.5% in February, minimum 63.2% in July). The proportion of annual PM2.5 and PM10 concentrations in Zabrze is unique among such coefficients for urbanized areas in the world: Azuza, China, has it equal to 0.68, Los Angeles, USA – 0.74, California, USA – 0.66, Montreal, Canada – 0.52, Toronto, Canada – 0.6.

In Zabrze, the coarse particles constitute about 13% of TSP on average, but in a few particular cases their contribution is over 30%. Average contribution of great particles, having the diameter greater than 10 μm, to TSP is 9.5% (Tab. 2).

Contribution of the coarse fraction to TSP increases in summer, when fine particle emission decreases due to heating cessation and coarse fraction, mainly of mechanical provenience, becomes more important (Figs 4 and 5).

Seasonally averaged diurnal cycle of PM1 mass concentrations in Leipzig, Germany, was described in [28]. The authors investigated mass concentration (computed from the number size distribution of particles) averaged for each of 24 hours of a day over the winter and summer separately. The average mass concentration for each winter hour of a day is much greater than for the same hour of a day in the summer. It very well reflects seasonal variability of PM1 concentrations. The situation is due to differences in meteorological conditions as well as nature of the particle emission.

In Zabrze, the PM1 concentration varied between 11.23 and 204.28 μg/m^3 in winter; on average it was 57.43 μg/m^3. In summer it was between 3.7 and 94.58 μg/m^3, the average being 25.13 μg/m^3 (Tab. 1).

In months April – September, the monthly average PM1 concentration was from 11.05 (August) to 47.69 (April) μg/m^3. For winter months the monthly PM1 average was from 35.12 (December) to 108.14 (January) μg/m^3. Despite of still lacking standard for

PM1, these values are alarming and exceed the PM10 standard. Importance of the problem may be appreciated by analyzing Figure 8.

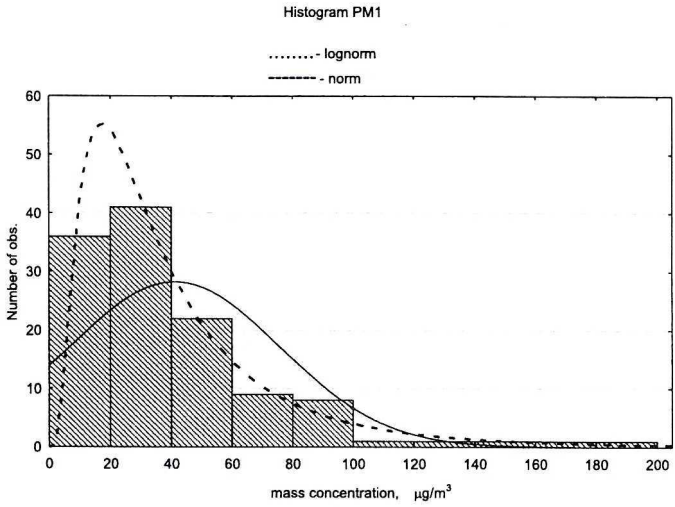

Fig. 8. Frequency distribution of the PM1 concentrations in Zabrze, Poland

Gravimetric measurements in Milan, Italy (urban conditions) showed summer PM1 content in PM2.5 at about 60%; in winter it was – 70% [27]. In 6 Australian towns, in winter, the PM1 contribution to PM2.5 was from 65 to 77% [14].

In Zabrze, contribution of PM1 to PM2.5 is higher than that reported in literature. In 2006, average contribution of PM1 to PM2.5 was 74.39% (Tab. 2) and there were many daily values almost reaching 90% (maximum – 92.2%). A very interesting thing was the average contribution of PM1 to PM2.5 in summer, equal to 77.7%, being higher than equal to 72.5% the winter contribution (Tab. 2). Such a result may be explained by an effect of far sources that additionally to the local ones affecting the site continuously during whole year or winter (combustion of fuels), contribute to pollution when winds blow from, for example, areas of high vehicular emission.

Figures 9 a–f present scatterplots for pairs of concentrations of a) PM2.5 versus PM1, b) PM2.5 versus PM1-2.5, c) PM2.5 versus PM10, d) PM10 versus PM1, e) PM10 versus PM1–2.5, f) PM10 versus PM2.5–10. Regression lines in 95% confidence interval are plotted and correlation coefficient r^2 is computed.

Regression coefficients are high if relation between coarser and comprised in it finer fractions is examined (e.g., r^2 for PM2.5 vs. PM1 is 0.95). PM2.5 and PM1–2.5 are much less correlated ($r^2 = 0.67$). The smallest correlation coefficient ($r^2 = 0.36$) was computed for PM10 vs. PM2.5–10, the highest – for PM10 vs. PM2.5 (0.99) and PM10 vs. PM1 (0.93), what may be in support of the above conclusion. The highest share in TSP the finest fractions had (Tab. 2, Figs 5 and 6) and this is a cause of high correlation between TSP and fine fraction concentrations. The correlations are so high that given the PM2.5 or PM1 concentration one can compute the PM10 concentration with very good adequacy (correlation is considered essential if $r^2 > 0.5$, for $r^2 > 0.8$ it is very good). Authors of [7, 14, 15] concluded their reasoning on scatterplots of component fractions

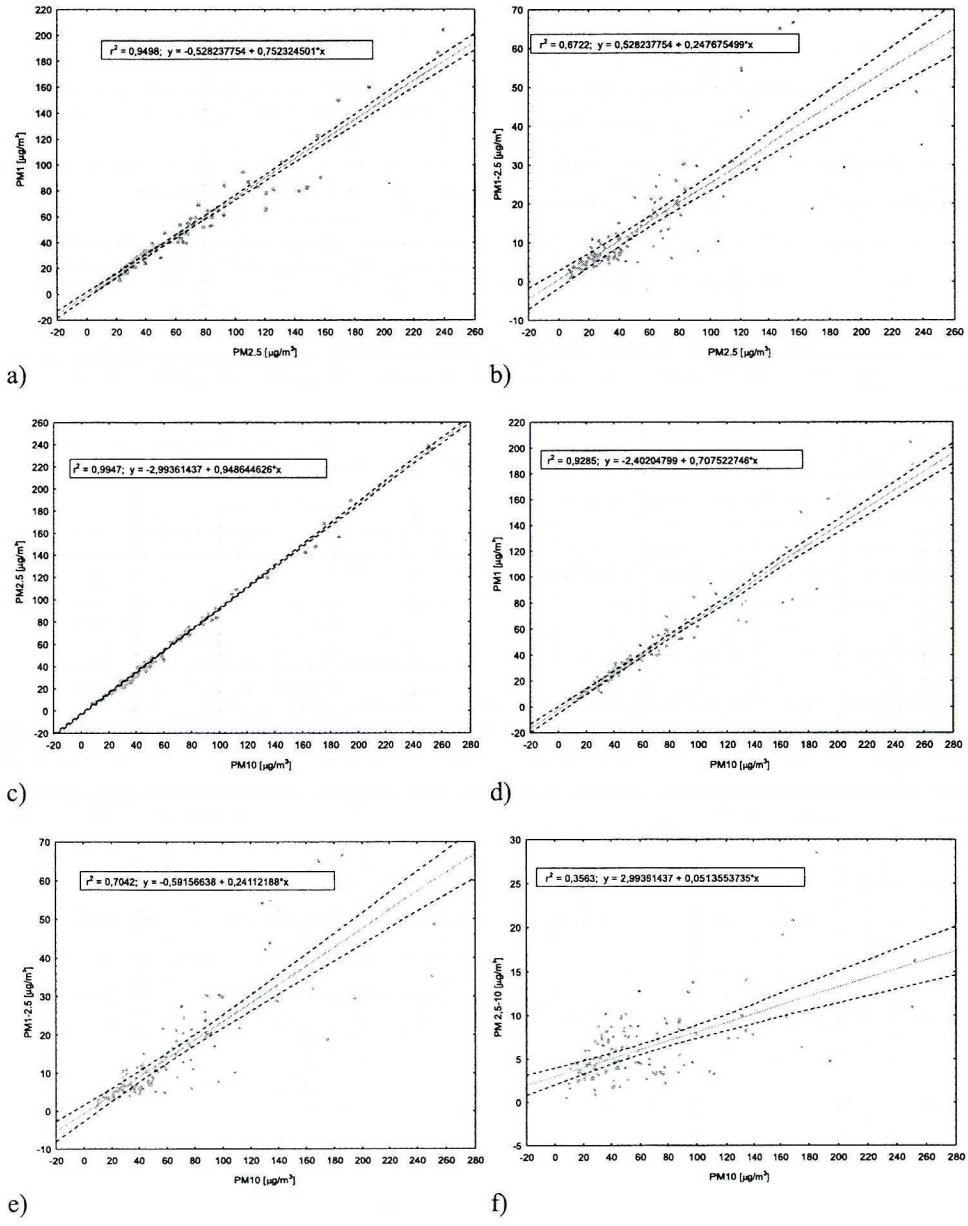

Fig. 9. Scatterplots of paired dust fraction concentrations

of PM10 similarly. The correlation coefficients for daily values of PM10 vs. PM2.5 concentrations in Bassel and Bern, Switzerland, measured in 1998–2001 were, 0.95 and 0.86 respectively [7], the correlation coefficient for PM1 vs. PM2.5 concentrations in Taipei, China, was 0.93 [7]. In the case of measurements at Australian various locations: Sydney (4 measurements), Brisbane (5 measurements), Melbourne (5 measurements), Canberra (3 measurements), Launceston (6 measurements), Adelaide (5 measurements), the cor-

relation coefficients for PM1 vs. PM2.5 and PM2.5 vs. PM10 concentrations were both equal to 0.98, while correlation coefficient for PM10–2.5 vs. PM10 was 0.35, although for PM2.5–1 vs. PM2.5 it was 0.94 [14].

CONCLUSIONS

In the Upper Silesian agglomeration ambient dust has been a crucial problem of air quality management. In spite of implementing the air protection program [6], the situation has not improved. Results presented in the paper evidence real possibility of violating existing and not reaching target air quality standards for dust, especially for PM2.5 (Tab. 1).

Analyses of seasonal concentrations and granulometric composition of ambient dust confirm significant share of dusts from combustion of fuels. High content of PM1 in TSP in the whole experimental period is especially alarming (Tab. 2). It means need to reformulate criteria evaluating actions undertaken to reduce emission of dust.

Results concerning PM1 in summer (Tab. 1 and Fig. 8) present contribution of far sources, e.g. vehicular, quite well. Higher contribution of dust particles of diameter greater than 2.5 μm in summer (Fig. 5) indicates occurrence of secondary emission from neighboring urban and agricultural terrains.

A high content of PM2.5 and PM1 in PM10 and TSP (Tab. 2) causes hazard to health of inhabitants within the investigated area. The exposure of people to the finest particles of dust is permanent – there is no technical method (such as ventilation) to remove the indoor finest particles (PM1). While the fine dust may be removed from the outdoor air by precipitation or wind, in living compartments, especially in winter, the finest dust particles may accumulate.

The obtained results justify a need for further investigations, especially determination of chemical composition of PM1.

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