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CONCENTRATIONS OF PM_{2.5}, PM_{2.5-10} AND PM-RELATED ELEMENTS AT TWO HEIGHTS IN AN URBAN BACKGROUND AREA IN ZABRZE (POLAND)

WIOLETTA ROGULA-KOZŁOWSKA, BARBARA BŁASZCZAK, KRZYSZTOF KLEJNOWSKI

Institute of Environmental Engineering PAS, 41-819 Zabrze M. Skłodowskiej-Curie str. 34 *Corresponding author's e-mail: wioletta@ipis.zabrze.pl

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Abstract: Concentrations and elemental composition of fine ($PM_{2.5}$) and coarse ($PM_{2.5-10}$) ambient particles, at two sampling points located at the same urban background sites, were investigated. The points were 20 m distant from each other and at various heights (2 and 6 m) above the ground. A dichotomous sampler, equipped with a virtual impactor, and a cascade impactor were used to sample the dust. An X-ray fluorescence spectrometer was used in the elemental analyses. The investigated urban background site. Coarse dust, whose concentration at 2.0 m above the ground was affected by secondary emission from roads, soil and other local low-level sources in some periods, appeared more heterogeneous.

INTRODUCTION

The need to measure ambient concentrations of particulate matter (PM) is due to the effects PM exerts on health, climate, and ecosystems. It arises from the necessity of identification of sources of PM and evaluation of effects of measures undertaken to limit ambient dust concentrations [6, 12, 13, 23, 62, 67]. In Europe, ambient PM is monitored by air quality monitoring stations (e.g. the *European Monitoring and Evaluation Programme* network, EMEP), which work within various domestic [1, 52] and international [6, 9, 13] projects. However, data on PM from various sites can be hardly compared or analyzed together because of different location criteria for the stations of their origin, limited size of the data sets [39], and, what is the most important, because of differences in PM samplers applied at different sites [24]. The domestic and international guidelines recommend the gravimetric method [5, 11, 45, 46, 62] as a standard for determination of the PM_{2.5} and PM₁₀ concentrations. However, the EU directives allow EU member countries to apply any method unless its consistency with the standard method cannot be proved.

Meteorological conditions [20, 32, 51, 52, 69], seasonal variability, and long-range transport of air pollutants [38, 52], but also the local conditions, affect strongly PM concentrations at a sampling site. Even very weak local sources may alter the regional representativeness of a sample [63]. The assumption of uniformity of $PM_{2.5}$, PM_{10} or $PM_{2.5-10}$ concentrations within particular urban area is usually forced by sparseness of measuring points (often a single measuring point within an area), but the actual dust concentrations in summer may vary from point to point even by 69% [23]. Addressing the issue, Wilson *et al.* (2005) [67] revue works concerning hazards from atmospheric aerosol which compare PM concentrations in different points of an urban area. On the one hand, they cite works that assume and base on the uniformity of the PM concentrations in an area, on the other hand, they cite works proving spatial variability of concentrations of all PM fractions. Concluding, they recommend an individual approach to every area.

Spatial distribution of $PM_{2.5}$ concentrations is relatively uniform. Higher concentrations may occur close to the sources. The $PM_{2.5\cdot10}$ concentrations are much more diversified. For example, PM_4 concentration at about 3 m above the ground may be lower by only 12% than at 1 m, while this difference is 35% for total PM [36]. The criterion of 1.5–4.0 m (or up to 8.0 m in special circumstances) above the ground for the height at which the sampling head of a dust sampler should be installed is clearly defined by the 2008/50/EC Directive [11]. In the context of [36], such optionality (1.5–8 m) allows methodology for measurement of concentrations of ambient dust, and consequently of the related elements, to have itself a serious effect on comparability of results. The goal of this work was to investigate concentrations and elemental composition of PM_{2.5} and PM_{2.5-10} at two, allowed by the 2008/50/EC Directive [11], heights (2 and 6m) at the same urban background site.

METHOD

The site selected for the experiment was an urban background area [11] in Zabrze. The PM concentrations at this site are representative of the concentrations in those living quarters of Upper Silesian cities that are affected by industrial and vehicular emissions.

At the first stage of the experiment, at two sites, horizontally distant by 20 m, samples of PM were taken with two simultaneously working samplers. The two samplers were: a sequential two-channel Ruprecht & Patashnik Dichotomous Partisol[®]-Plus Model 2025 (Partisol) [31] and a three-stage Dekati[®]PM₁₀ cascade impactor (Dekati) [29]. The sampling head of Partisol was located 2.0 m above the ground, the sampling head of Dekati—6.0 m. Dekati was located on the roof of the Institute building (Figure 1). On every sampling day, at 12:00 midnight (Table 1), both samplers started simultaneously and were working for 48 hours – Decati continuously, Partisol automatically changed its substrate filter after 24 hours.

This allowed for collection of sufficient amount of PM on each Dekati stage and prevented the filters in Partisol from overloading. Measurements of PM concentrations were accompanied by measurements of basic meteorological parameters (Table 1, the Regional Inspectorate for Environmental Protection automatic station, located 20 m from Partisol and 40 m from the Institute building – Figure 1).

Dariad	Height	Т	Р	V	RH	Pr.	PM ₂₅	PM _{2 5-10}	PM ₁₀
Period		$[^{\circ}C]^{a)}$	[hPa] ^{b)}	$[m \cdot s^{-1}]^{c)}$	[%] ^{d)}	[mm] ^{e)}	[µg·m ⁻³]	[µg·m ⁻³]	[µg·m ⁻³]
26 - 27.04.2008	6 m ^{f)}	0.5	002	0.75	(0.5	0	39.2	4.1	43.3
	2 m ^{g)}	9.5	992	0.75	09.3	0	48.8	12.7	61.5
29 - 30.04.2008	6 m	11.05 072 1.15 (5.5 0	30.0	7.0	37.0				
	2 m	11.05	975	1.15	03.3	U	32.3	41.9	74.2
07 - 08.05.2008	6 m	10.45	0.96		0	27.8	6.1	33.9	
	2 m	10.45	980	0.9	07	0	29.1	17.8	46.9
14 - 15.05.2008	6 m	14	070.5	0.55	565	0	31.9	9.2	41.1
	2 m	14	9/9.5	0.55	30.3	0	22.5	17.0	39.5
28 - 29.05.2008	6 m	14.6	985	1.7	52.5	0	26.4	5.6	32.0
	2 m						13.5	18.2	31.7
04 - 05.06.2008	6 m	15 55	080	1.4	72	2.0	17.1	5.2	22.3
	2 m	13.33	980	1.4	/3	5.9	17.3	14.0	31.3
11 - 12.06.2008	6 m	6 m 15 4		0.6	02	4.4	18.9	5.7	24.6
	2 m	13.4	9/9	0.0	92	4.4	19.4	14.0	33.4
18 - 19.06.2008	6 m	16.25	002 5	0.7	(5.5	0	26.6	6.5	33.1
	2 m	10.23	982.3	0.7	03.3	0	25.7	14.6	40.3
25 - 26.06.2008	6 m	10.05	0.02	0.6	76.6	1.5	22.8	7.0	29.8
	2 m	19.05	983	0.0	13.5	1.5	21.9	15.6	37.5
02 - 03.07.2008	6 m	21.4	000	0.5	12.5	0	30.6	9.0	39.6
	2 m	21.4	900	0.5	43.3	U	25.9	20.1	46.0

Table 1. Average 48h weather conditions and concentrations of fine (PM_{2.5}) and coarse (PM_{2.5-10}) dust at two heights (2 and 6 m) in Zabrze in 26.04–03.07.2008

*) weather conditions for the site were taken from the RIEP Internet site; they were used to compute two-day averages of meteorological data (arithmetical means for two days)

^{a)} T – air temperature; ^{b)} P – atmospheric pressure; ^{c)} V – wind speed at 14 m above the ground level; ^{d)} RH – relative humidity of air; ^{e)} Pr. – precipitation, ^{f)} sampled with Dekati, ^{g)} sampled with Partisol



Figure 1. Arrangement of the samplers at the measuring site in Zabrze: Dekati, Partisol, and TEOMs (6, 2, 4 m above the ground, respectively)

The experiment was carried out in spring and summer, between 26 April and 3 July, because in winter very high municipal emission obscures contributions of other sources (e.g. vehicular) in the area [44, 53].

For both, $PM_{2.5}$ and $PM_{2.5-10}$ 48h concentrations were determined from masses of collected dust and volumes of air passed through the samplers. The masses were de-

termined gravimetrically, according to [45, 46]. The substrates were conditioned for 48 hours in the weighing room (air humidity $45 \pm 5\%$, air temperature $20 \pm 2^{\circ}$ C) before weighing. Each substrate was weighed twice with a 24 h period between the weighings, before and after exposure, on a Mettler Toledo microbalance (resolution 2 µg). A clean substrate was rejected if its two weights differed more than 10 µg. Before chemical analyses, substrates were stored in tight containers in a refrigerator.

All samples were analyzed for their elemental composition by using energy dispersive X-ray fluorescence (EDXRF). PANalytical Epsilon 5, used in the analyses, was equipped with a water-cooled X-ray tube (Gadolinium anode, working range 25–100 kV, 150 μ m thick Be side window), system of nine secondary targets (Al, Ti, Fe, Ge, Zr, Mo, Ag, Ce₂O₃, Al₂O₃), Ge(Li) detector (resolution of 140 eV, energy range 0.7–100 keV, 30 mm² active area, 8 μ m thick Be window).

Measurements were done in vacuum, each analysis lasted for 4800 s (the whole runtime of the analytical program using secondary targets: Al, Ti, Fe, Ge, Zr and Al_2O_3). The concentrations of particular analytes were determined by comparing the results with calibration curves. The curves were determined by applying the Micromatter Inc. thin layer standards, the factors correcting the inter-elemental effects were referred to.

The limits of detection and recoveries of the standard (NIST 2873) for the procedure are presented in Table 2.

Flamont	Limit of	detection	Pecovery %
Liement	ng·cm ²	ng∙m ^{3*}	Recovery, 70
S	1.5	0.7	96
Cl	1.3	0.6	-
Ca	1.2	0.6	86
Ti	0.8	0.4	99
Cr	0.9	0.4	103
Fe	0.7	0.3	102
Ni	0.6	0.3	88
Zn	0.9	0.4	112
As	11.6	5.8	84
Br	0.4	0.2	-
Cd	4.5	2.2	-
Pb	0.3	0.1	116

Table 2. Limits of detection and recoveries of the standard for the EDXRF procedure for determination of content of elements in suspended dust collected on Teflon filters

* flow rate 21.6 m3

RESULTS

Mass concentrations of PM_{2.5} and PM_{2.5-10}

In Table 1, 48 h concentrations of $PM_{2.5}$ and $PM_{2.5-10}$ are presented. Concentrations were measured with the use of two samplers, Partisol and Dekati, at the same urban background site in Zabrze, at two different heights: 2.0 m (Partisol) and 6.0 m (Dekati) above the ground. Dekati allows for simultaneous measurement of concentrations of $PM_{1,2}$

 $PM_{1.2.5}$, and $PM_{2.5-10}$. The Dekati-measured $PM_{2.5}$ concentration is the sum of the concentrations of dust collected on the Dekati afterfilter and its third stage $(PM_1+PM_{1.2.5})$. The average concentrations of $PM_{2.5}$ in the measuring period (26.04–03.07.2008) were 26 μ g·m⁻³ at 2.0 m and 27 μ g·m⁻³ at 6.0 m. The minimum $PM_{2.5}$ concentrations, 14 μ g·m⁻³ at 2.0 m and 17 μ g·m⁻³ at 6.0 m, occurred on 28–29.05 and 04–05.06, respectively (Table 1). The maxima, 49 μ g·m⁻³ at 2.0 m and 39 μ g·m⁻³ at 6.0 m, occurred on 26–27.04. Also on 26–27.04, the lowest $PM_{2.5-10}$ concentrations, 13 μ g·m⁻³ at 2.0 m and 4 μ g·m⁻³ at 6.0 m, occurred. The maximum $PM_{2.5}$ concentrations are much higher than the other $PM_{2.5}$ concentrations.

The concentrations of $PM_{2.5-10}$, like those of $PM_{2.5}$, fell into a very wide interval. The average $PM_{2.5-10}$ concentration at 2.0 m was 19 µg·m⁻³, almost three times the average concentration at 6.0 m (7 µg·m⁻³, Table 1). The maximum $PM_{2.5-10}$ concentration at 6.0 m, 9 µg·m⁻³, occurred twice: on 14-15.05 and 02–03.07; the one at 2.0 m, four times greater (42 µg·m⁻³), occurred on 29–30.04.

Several independent factors shape concentrations of air pollutants. The most effective are emission, meteorological conditions, and geographical location [33, 49, 52]. Nevertheless, in the period 26.04–03.07.2008, neither fine nor coarse dust concentrations were significantly correlated with the meteorological parameters (Table 3).

	Partisol	Dekati	TEOM ^{a)}	$T [°C]^{b)}$	P [hPa] ^{c)}	$V [m/s]^{d}$	RH [%] ^{e)}	Pr. [mm] ^{f)}			
	PM ₂₅										
Partisol ^{g)}	1	0.78	0.90	-0.41	0.40	-0.25	-0.51	-0.81			
Dekati ^{h)}	-	1	0.74	-0.55	0.42	-0.33	0.01	-0.43			
TEOM ⁱ⁾	-	-	1	-	-	-	-	-			
	PM _{2 5-10}										
Partisol	1	0.28	-	0.51	-0.54	-0.47	-0.54	-0.33			
Dekati	-	1	-	-0.23	-0.66	0.23	-0.22	-0.32			
TEOM	-	-	1	-	-	-	-	-			
PM_{10}											
Partisol	1	-	0.51	-	-	-	-	-			
Dekati	-	1	0.79	-	-	-	-	-			
TEOM	_	_	1	_	_	_	_	_			

Table 3. Pearson linear correlation coefficients for sets of 48h average concentrations determined gravimetrically (PARTISOL/DEKATI) and automatically (TEOM) and Pearson linear correlation coefficients for meteorological parameters and concentrations of PM_{2.5} and PM_{2.5-10} in 26.04–03.07.2008

^{a)} TEOM for PM₂₅ measurements was operated by the authors of the paper; TEOM for PM₁₀ measurements was operated by the Regional Inspectorate for Environmental Protection;

^{b)} T – air temperature; ^{c)} P – atmospheric pressure; ^{d)} V – wind speed at 14 m above the ground level; ^{e)} RH – relative humidity of air; ^{f)} Pr. – precipitation, ^{g)} sampled at 2m, ^{b)} sampled at 6m, ⁱ⁾ sampled at 4m

Possibly, the insufficiency of data prevented the credible statistical analysis. Relatively strong linear correlations were found between precipitation and $PM_{2.5}$ concentrations (Pearson correlation coefficient r = -0.81), and between atmospheric pressure and $PM_{2.5-10}$ concentrations (r = -0.66). The lack of correlations between the concentrations and majority of meteorological parameters may also be due to domination of local dust sources [49].

The 48h concentrations of $PM_{2.5}$ at both heights were fairly well correlated (r = 0.78 – Table 3). However, the differences between them were significant (even 13 µg·m⁻³) on 28–29.05 (Table 1). Except for the first three pairs, where the concentrations differed by at most 10 µg·m⁻³ on 26–27.04, the concentrations at 6.0 m were higher than or (like on 04–05.06 and 11–12.06) equal to the concentrations at 2.0 m (Table 1).

The differences between $PM_{2.5-10}$ concentrations at 2.0 m and 6.0 m were between 7.8 μ g·m⁻³ (14–15.05) and 34.9 μ g·m⁻³ (29–30.04). Their average, 12 μ g·m⁻³, is three times higher then the average difference between the $PM_{2.5}$ concentrations. The Pearson coefficient (r = 0.28, Table 3) proves a lack of correlation between $PM_{2.5-10}$ concentrations at the two heights. The spatial variability of $PM_{2.5-10}$ concentration is considerable in the area.

The results from Partisol and Decati were also checked for their linear correlation with the results from automatic monitors of PM_{10} and $PM_{2.5}$ (TEOMs). The TEOM station was located about 20 m from Partisol and 40 m from Decati. The TEOM measuring $PM_{2.5}$ was operated by the authors, whereas the TEOM measuring PM_{10} was operated by the Regional Inspectorate for Environmental Protection (RIEP) in Katowice [68]. The sampling heads of the two TEOMs were at 4 m above the ground level.

The PM_{2.5} concentrations from Partisol were better correlated with the PM_{2.5} concentrations from TEOM (r = 0.67) than the concentrations from Dekati. There was no significant correlation between the results from Partisol and TEOM for PM_{2.5-10}; the linear correlation coefficient for PM_{2.5-10} concentrations from Dekati and TEOM was r = 0.79(Table 3). It means that in the period 26.04–03.07.2008 the height of sampling head had a great effect on measurement of concentration of dust, especially coarse dust. Obviously, the Partisol head, 2.0 m above the ground, collected secondary dust.

The Decati and TEOM heads were located at heights excluding or strongly limiting effects of secondary $PM_{2.5-10}$ emission. This explains as well the good linear correlation of the $PM_{2.5-10}$ concentrations from Dekati and TEOM as the Partisol-Dekati and the Partisol-TEOM differences in these concentrations. Because secondary emission adds almost exclusively to coarse particles [57] the difference in heights of sampling heads did not affect significantly the $PM_{2.5}$ concentrations.

The better correlation between $PM_{2.5}$ concentrations from Partisol and TEOM is probably due to identity of their sampling heads (US EPA [61], air flow rate 1 m³·h⁻¹). Concentrations from longer periods covering uniformly winter (slighter effect of secondary emission on $PM_{2.5-10}$ concentrations) and summer are in high linear correlation as well when measured with Partisol and Decati as with Partisol and TEOM for both $PM_{2.5}$ and $PM_{2.5-10}$ [30].

The results from the period 2007–2008 from the investigated site are presented in Table 4.

Partisol was used to measure 24 h concentrations in uneven months of 2007 and the whole year 2008. In 2007–2008 Dekati started every third day and measured 48 h concentrations. The $PM_{2.5}$ concentrations from 04–07.2008 (this study) are lower than the yearly average concentrations, the $PM_{2.5-10}$ concentrations from 04–07.2008 are higher than the averages from 2007 and 2008 (Table 4).

In general, the $PM_{2.5-10}$ concentrations show seasonal fluctuations. They assume higher values in spring (greater part of 04–07.2008) and winter, lower in summer and autumn [17]. High yearly $PM_{2.5}$ concentrations are caused by high concentrations in winter (heating season) – not only in the south of Poland (Zabrze) but also in other European

Country	Location, measuring period	PM _{2.5}	PM _{2.5-10}
		[µg·m ⁻ ']	[µg·m ⁻³]
	Zabrze (urban background, PARTISOL); IV-VII 2008 a)	27	19
	Zabrze (urban background, DEKATI); IV-VII 2008 a)	26	7
Poland Upper	Zabrze (urban background, PARTISOL); 2007 ($n = 181$) ^{b)}	33.1	10
Silesia	Zabrze (urban background, PARTISOL); 2008 ($n = 354$) ^{b)}	40.5	9
	Zabrze (urban background, DEKATI); 2007 ($n = 106$) ^{b)}	38.5	5
	Zabrze (urban background, DEKATI); 2008 $(n = 81)^{b}$	42.7	5
	Zabrze(urban background station);smog episode (01.2006) ^{c)}	187.30	25.19
Finland	Helsinki (urban background); 23 VIII – 23 IX 2002 ^d)	11.5	6.82
	Tarragona (urban background); 2001 ^{e)}	21.8	15.6
	Huelva (urban background); 2001 ^{e)}	19.3	18.2
Spain	Alcobendas (urban background); 2001e)	24.9	7.3
	Llodio (urban background); 2001r. e)	23.9	7.8
	Madrid (urban background); 1999 – 2000e)	34.1	13.6
Netherlands	Arnhem (background); VIII – IX 1994 ^{g)}	35.0	26.8
Scotland	Fort William (suburban station); 2007 ^{h)}	10	4
Gautian	Zurich-Kaserne (city center, yard); IV 1998r III 1999r. i)	20	6.3
SWISS	Basilea (suburb); IV 1998r. – III 1999 ⁱ)	18.9	6.7
	Erzurum; II 2005r. – II 2006r. ^{j)}	12.67	17.96
Turkey	Erzurum; heating season ^{j)}	19	17
	Erzurum; non-heating season ^{j)}	6	19
Southern	Port Talbot (industrial station); 26 IV – 25 V 2001 ^k)	83.61	54.73
Wales	Cardiff (urban station); $22 I - 06 III 2001^{k}$	89.87	26.31
Hungary	Budapest (urban background); 12 – 17 IV 1999 ¹⁾	30	35
Hungary	Budapest (city centre, park); 19 IV – 13 V 1999 ¹⁾	28	41
. 1	Milan (urban station); winter 1997/1998 ^{m)}	70	40
Italy	Milan (urban station); summer 1998 ^{m)}	51	23
Mexico	Mexico city (urban area, various sources); III 2006 ⁿ⁾	50	37.7
Brazil	Rio de Janeiro; X 1998r. – IX 1999°)	17.1	25.7
	Brisbane; II – IV 2003r. ^{p)}	4.5	7.3
Australia	Melbourne (Australia); II – IV 2003 ^{p)}	5.9	8.3
	Sydney (Australia); II – IV 2003 ^{p)}	5.5	8.3
Southern Africa	Qualabothia (urban area); winter 1997 ^{q)}	115.46	11.95
	Dhaka (living quarter, moderate traffic); I – IV 2006 ^{r)}	63.6	10.1
Bangladesh	Dhaka (urban-rural area): $I - IV 2006r.$ ^{r)}	57.1	4.9
China	Guangzhou (city center, dense traffic): 02 VIII – 10 IX 2004 ^{s)}	103.35	41.17
India	Kolkata (agglomeration): winter 2002 th	178.57	125.47
Lebanon	Beirut (living and commercial area): III – VIII 2004^{u}	33.50	54.03
	Beirut (living and commercial area): II $2004r - I 2005^{u}$	34.36	64.96
Taiwan	Taichung Harbor (spring– III IV V) $^{(v)}$	49.3	37.0
	Taichung Harbor (summer – VI VII VIII) ^{v}	46.4	26.6
	Taichung Harbor (autumn – IX X XI) ^{v}	43.0	26.2
	Taichung Harbor (winter – XII D^{ν})	49.5	36.0
Vietnam	Ho Chi Minh (urban station) ^{w)}	32	16

Table 4. Mass concentrations of $PM_{2.5}$ and $PM_{2.5-10}$ at selected sites in Europe and the world

^{a)} [this study]; ^{b)} [investigation conducted in IEE PAS]; ^{c)} [44]; ^{d)} [58]; ^{e)} [48]; ^{f)} [47]; ^{g)} [26]; ^{h)} [1]; ⁱ⁾ [26]; ^{j)} [3]; ^{k)} [38]; ^{l)} [57]; ^{m)} [34]; ⁿ⁾ [40]; ^{o)} [35]; ^{p)} [22]; ^{q)} [14]; ^{f)} [55]; ^{s)} [64]; ^{t)} [10]; ^{u)} [56]; ^{v)} [17]; ^{w)} [16]

(e.g. Italy) and world (The Republic of South Africa, Turkey, Taiwan, India) urbanized areas (Table 4). Accidentally, intensive combustion of fossil fuels in heating season, yielding rapidly peaking emission of fine dust from low-level domestic sources, together with specific weather conditions, cause smog episodes similar to the one that occurred in Zabrze in January 2006 (Table 4).

PM_{2.5} concentrations at urban background sites in Finland, Scotland, Switzerland, the Netherlands, Brazil or Llodio in Spain are lower than in Zabrze. But in many areas in Europe (Barcelona in Spain, Arnhem in The Netherlands, Hungary, Italy, Southern Wales), North America (Mexico), Africa (The Republic of South Africa), Asia (Bangladesh, China, Lebanon, Taiwan, Vietnam), PM_{2.5} concentrations are significantly higher (Table 4).

Only at few sites listed in Table 4 $PM_{2.5-10}$ concentrations are lower than the concentrations at 6.0 m above the ground in Zabrze (Helsinki, Fort William, Zurich-Kaserne, Basel). Majority of the $PM_{2.5-10}$ concentrations from almost the whole world are higher than the concentrations in Zabrze in the considered period (Tables 1, 4). The difference between yearly average $PM_{2.5-10}$ concentration in Zabrze (in 2007 and 2008, especially measured at 6.0 m with Dekati) and the concentrations in other regions is yet more distinctive (Table 4). It means great, much greater than in other sites listed in Table 4, contribution of $PM_{2.5}$ to PM in Zabrze. Exceptionally high carbon content indicates anthropogenic origin of fine ambient dust in Zabrze [43].

The differences in the concentrations, especially of $PM_{2.5-10}$, occur for relatively little differing heights above the ground (4 m). Although the results, received at two distant by 20 m points with two different samplers within a short period of time, do not allow for quantitative determination of the effect of height of a head location on the measurements, it is obvious that the effect is significant to monitoring of air quality, especially for PM_{10} . The differences between yearly means of daily concentrations, of both $PM_{2.5-10}$, at the two points are small (Table 4). It means that the big differences, observed in 04–07.2008 (especially for $PM_{2.5-10}$), are neither due to the distance nor to the construction of the samplers but to the different heights of the heads location. It also means that this height difference is not probable to cause so big a difference in yearly concentrations as to occur exceeding of the standard at one height and not at the other, but the episodes of elevated $PM_{2.5-10}$ concentrations, such as described in this study, may cause violation of the standard for 24 h PM_{10} concentrations (50 µg·m⁻³, [11]) at only one height.

In 26.04–03.07.2008, concentrations of PM₁₀ exceeded the standard on 26–27.04 and 29–30.04 with the values 62 μ g·m⁻³ and 74 μ g·m⁻³, respectively, but it was indicated only by Partisol, 2.0 m above the ground. In such a short period the standard for PM₁₀ was exceeded twice, therefore, the risk of exceeding the standard more often than 35 times in a year is high. The fact is significant because people inhale air from the height of 2.0 m above the ground level. Coarse dust occurring in such high concentrations may adversely affect human health [12, 15, 28, 41, 54].

Elemental composition of PM_{2.5} and PM_{2.5-10}

Because of different origins and times of persistence in the atmosphere [21], $PM_{2.5}$ and $PM_{2.5,10}$ significantly differ in the elemental composition (Figure 2).







Among the PM_{2.5}-related elements, at 2.0 m above the ground (Partisol-measured), iron had the greatest average 48h ambient concentration ($C_{av} = 402.6 \text{ ng} \cdot \text{m}^{-3}$); next were sulfur ($C_{av} = 383.8 \ \mu\text{g} \cdot \text{m}^{-3}$) and calcium ($C_{av} = 105.0 \ \text{ng} \cdot \text{m}^{-3}$). The average concentrations of the rest of the elements were lower than 100 ng·m⁻³ – the concentrations of nickel ($C_{av} = 0.6 \ \text{ng} \cdot \text{m}^{-3}$), cadmium ($C_{av} = 1.9 \ \text{ng} \cdot \text{m}^{-3}$), and arsenic ($C_{av} = 2.7 \ \text{ng} \cdot \text{m}^{-3}$) were lowest. All the elements, except bromine and cadmium, were almost entirely contained in PM_{2.5-10}. The ambient concentrations of some (especially crustal) PM_{2.5-10}-related elements were drastically higher than the concentrations of the PM_{2.5}-related ones: average concentration of PM_{2.5-10}-related calcium was 5442.5 ng·m⁻³ while the concentration of calcium from PM_{2.5} was 105.0 ng·m⁻³ (Figure 2). It was 7491.8 ng·m⁻³ and 402.6 ng·m⁻³, respectively, for iron.

The opposite is true of dust sampled at 6.0 m above the ground (Dekati) except for calcium, the greater amount of each element was accumulated in PM_{2.5}. Among the PM_{2.5}-related elements, iron ($C_{av} = 794.5 \text{ ng} \cdot \text{m}^{-3}$), sulfur ($C_{av} = 537.4 \text{ ng} \cdot \text{m}^{-3}$), calcium ($C_{av} = 263.7 \text{ ng} \cdot \text{m}^{-3}$), and zinc ($C_{av} = 106.4 \text{ ng} \cdot \text{m}^{-3}$) had the greatest ambient concentrations; among the PM_{2.5-10}-related elements, iron ($C_{av} = 471.0 \mu \text{g} \cdot \text{m}^{-3}$) and calcium ($C_{av} = 276.6 \text{ ng} \cdot \text{m}^{-3}$). Except for chlorine and sulfur ($C_{av} = 56.9 \text{ ng} \cdot \text{m}^{-3}$ and $C_{av} = 36.1 \text{ ng} \cdot \text{m}^{-3}$, respectively), the rest of the 48h ambient concentrations of the PM-related elements were not greater than 30 ng $\cdot \text{m}^{-3}$.

Except for arsenic and cadmium, whose average ambient concentrations at both heights were close, the concentrations of almost all $PM_{2.5}$ -related elements were about two times greater at 6.0 m than at 2.0 m. Instead, the concentrations of $PM_{2.5-10}$ -related elements at 2.0 m were several times greater than at 6.0 m. Because the head of Dekati was placed 4 m higher than the head of Partisol, it suggests the effect of a low-level emission source [42].

Among the $PM_{2.5}$ -related non-metals, sulfur had the greatest ambient concentrations; among the $PM_{2.5-10}$ -related non-metals it was chlorine. Greater part of PM-related sulfur at 2.0 m was in $PM_{2.5}$ (Figure 2). Some authors maintain that sulfur in ambient fine dust originates mainly from combustion of fossil fuels [7] and photochemical reactions in the atmosphere [7, 42]. High ambient concentrations of $PM_{2.5}$ -related sulfur at the two heights are due to the origin of $PM_{2.5}$ from these sources.

PM-related chlorine may originate from various sources but in an urban area it is mostly anthropogenic. High ambient concentrations of $PM_{2.5}$ -related chlorine are probably due to combustion of, among others, coal and wastes. Exceptionally high, much higher than the $PM_{2.5}$ -related, concentrations of $PM_{2.5-10}$ -related sulfur and chlorine at 2.0 m may reflect the effect of re-suspended road dust. Chlorine, together with many other elements, in PM_{10} may originate from the abrasion of car breaks [18].

Among crustal elements, calcium, titanium, and iron [2], iron had the highest ambient concentrations. Most of iron in PM at 2.0 m was in $PM_{2.5-10}$ (Figure 2). Similar results, about 3.5 times higher the $PM_{2.5-10}$ -related than the $PM_{2.5}$ -related iron concentrations, are reported by [16]. At 6.0 m, iron occurred rather in $PM_{2.5}$.

Iron is one of the most abundant elements in the Earth crust, therefore, high iron content of PM may prove the mineral origin of a great part of the investigated PM [2]. On the other hand, many stationary sources and industrial processes emit great amounts of iron [19, 50]. It may give an explanation for the presence of iron in PM_{2.5}, especially at 6.0 m above the ground, beyond any effects of secondary emission. High ambient concen-

trations of calcium and titanium are commonly attributed to the propagation of mineral dust and, in the case of calcium, of abrasive dust from road surface and soil dust [42]. It agrees with the high $PM_{2.5-10}$ -related calcium and titanium concentrations at 2.0 m, higher than the concentrations of calcium and titanium from $PM_{2.5}$ and from both PM fractions at 6.0 m.

Except for arsenic and cadmium, the concentrations of $PM_{2.5}$ -related heavy metals were higher at 6.0 than at 2.0 m (Figure 2). Like sulfur, they may indicate high contribution of combustion of fossil fuels and energy production-related sources [42]. Moreover, almost all heavy metals listed in Figure 2 are related to $PM_{2.5}$ from combustion of gasoline and diesel oil in cars [65].

Like the other elements, the $PM_{2.5-10}$ -related heavy metals had higher, often several times, concentrations at 2.0 m. It confirms the earlier assumption that PM from roads has great contribution to coarse dust in the considered period.

To discriminate between crustal (natural) and anthropogenic elements in PM, the element enrichment factors (*EF*) in PM_{2.5} and PM_{2.5-10} were analyzed [8, 37, 50]. The enrichment factor EF_y for an element x is defined as:

$$EF_{x} = \frac{(C_{x} / C_{ref})_{PMi}}{(C_{x} / C_{ref})_{crust}}$$
(1)

where PM_i is $PM_{2.5}$ or $PM_{2.5-10}$, C_x and C_{ref} designate concentrations of the element x and the reference element, $(C_x/C_{ref})_{PMi}$ and $(C/C_{ref})_{crust}$ are proportions of these concentrations in PM_i and in the Earth crust, respectively. The observed concentrations C_x were referred to the concentration C_{Ti} of titanium, a marker element for the Earth crust. Consequently, $EF_{ti} = 1$.

The data on the chemical composition of the upper continental crust were taken from the literature [66]. EF_x close to 1 means crustal origin of the element x; high EF_x means other (generally anthropogenic) origin of x [4, 27, 37]. Because the elemental composition of the local Earth's surface differs from the averaged composition of the upper continental crust the anthropogenic origin was attributed to the elements x with $EF_x > 10$.

EFs, computed for twelve elements in $PM_{25,10}$, and $PM_{25,10}$, are presented in Table 5.

All the elements in Table 5 may be divided into two groups. The first group comprises the elements with low EF ($EF \le 10$), falling between $EF_{Ca} = 1.4$ and $EF_{Fe} = 5.7$ (Ca and Fe PM_{2.5}-related, Partisol-measured), i.e. elements with the probable natural origin (soil dust). Because of low EF, the crustal origin might be attributed to PM_{2.5-10}-related sulfur from Dekati and to PM_{2.5-10}-related chlorine and chromium from Partisol. However, despite their low EFs their other important source may be fly ash [60].

The second group comprises the elements having high *EF*s both in $PM_{2.5}$ and in $PM_{2.5-10}$. It comprises Cd, Br, Pb, Zn, As with very high *EF*s, and S, Cl, Cr, Ni with lower ones. All heavy metals, except Cr and Ni, have *EF*s greater than 100. Therefore, because of high *EF*s, these elements are of anthropogenic origin in both $PM_{2.5-10}$. They probably come from combustion of hard coal for heating, combustion of wastes, high-temperature industrial processes [49, 64], vehicular sources (car exhaust, re-suspended road dust, particles from abrasion of tires and breaks, particles from other parts of cars) [25, 44, 59].

The value of EF_x depends on the dust fraction the element x is related to and the location of a sampler (Table 5).

Ele	ement	S(f) ^{a)}	$S(c)^{b)}$	C1(f)	C1(c)	Br(f)	Br(c)
Mean		104.8	14.0	20.2	0.6	1337.7	14.0
2m ^{c)}	D	194.0	14.0	29.2	9.0	1337.7	14.0
	Range	43.5÷4/2.1	3.8÷49.7	0.8÷57.6	6.0÷13.3	97.4÷3246.9	0.0÷40.9
6md)	Mean	115.2	8.3	22.1	19.9	714.2	0.0
	Range	31.5÷427.1	5.0÷24.9	5.8÷87.0	5.4÷61.4	278.3÷2783.0	0.0
Elem	ent	Ca(f)	Ca(c)	Ti(f)	Ti(f) Ti(c) Fe(f)		Fe(c)
2	Mean	1.4	1.7	1.0	1.0	5.7	2.2
2111	Range	0.5÷2.3	1.2÷2.2	1.0	1.0	1.7÷17.4	1.7÷3.2
6.00	Mean	1.4	2.0	1.0	1.0	4.4	3.3
om	Range	0.9÷2.4	1.1÷5.5	1.0	1.0	2.1÷8.0	1.7÷10.1
Element		Cr(f)	Cr(c)	Ni(f)	Ni(c)	Zn(f)	Zn(c)
2m	Mean	46.9	10.3	18.7	2.6	566.6	89.7
	Range	4.5÷77.9	6.2÷15.5	0.0÷55.9	0.0÷7.8	110.9÷1043.0	34.2÷172.5
6m	Mean	21.8	14.3	14.1	5.6	367.9	121.4
om	Range	7.6÷56.3	0.0÷89.1	4.8÷47.9	0.0÷33.5	162.7÷1019.0	0.0÷383.6
Element		As(f)	As(c)	Cd(f)	Cd(c)	Pb(f)	Pb(c)
2m	Mean	594.8	154.7	9829.8	336.8	1079.2	124.0
	Range	0.0÷1558.5	18.2÷419.3	0.0÷25465.7	0.0÷3216.7	210.9÷2230.7	42.3÷203.8
6m	Mean	262.0	0.0	3626.0	648.2	616.3	193.0
	Range	0.0÷1558.5	0.0	0.0÷13096.6	0.0÷3395.4	237.7÷1702.6	42.3÷623.4

Table 5. Enrichment factors for selected elements in $PM_{2.5}$ (f) and $PM_{2.5-10}$ (c) sampled at two heights (2 and 6m) in Zabrze

a) (f)-fine dust (PM2,) b) (f)-coarse dust (PM2, 10) c) sampled with Partisol, d) sampled with Dekati

All the elements, except Ca, have *EFs* higher, often several times, in PM_{2.5} than in PM_{2.5-10}. It suggests the anthropogenic origin of fine dust at both heights and agrees with other studies on urban PM [26, 34, 48]. The contribution of natural sources to the emission of PM_{2.5} is small. Moreover, the differences between EF_x in PM_{2.5} and EF_x in PM_{2.5-10} are much smaller in the first than in the second group.

EFs of the elements of natural origin were similar at both heights, although almost all slightly higher at 6.0 m (except S in $PM_{2.5-10}$ and Fe in $PM_{2.5}$). *EFs* of majority of anthropogenic elements in $PM_{2.5}$ were greater at 2.0 m; *EFs* of majority of anthropogenic elements in $PM_{2.5-10}$ —at 6.0 m.

The analysis of *EF*s provides a good confirmation of high spatial variability of the elemental composition of both PM fractions. The effect of re-suspended road dust, a mixture of combustion [59] and non-combustion [25, 59, 65] particles from vehicular sources on concentrations of, especially coarse dust-associated, elements at the lower height is clear. However, the presence of heavy metals in coarse dust may also be due to the pollution from soil or other components of the environment [57]. Road traffic and movements of air raise deposited pollutants back into the atmosphere [19, 44]. The road dust particles are permanently made to move by passing cars and the finer particles stay suspended in the air [60]. The ambient elements from road dust may be accumulated as well in fine as in coarse dust.

Road dust is a considerable source of suspended dust and the associated elements, including heavy metals, within many urban areas [19, 59]. The emission from low-level

sources (car exhaust and road dust) may cause great spatial differentiation of concentrations and elemental composition of ambient dust. However, the contribution of road dust to ambient dust depends on a season of a year and meteorological conditions [7]. In the periods of heavy rain or snow, the differences in concentration and elemental composition of coarse dust may not be as great as presented here or they may be even negligible.

CONCLUSION

The ambient concentrations and elemental composition of $PM_{2.5}$ and $PM_{2.5-10}$ at the urban background site in Zabrze in spring and summer strongly depend on the height above the ground level, those of the $PM_{2.5-10}$ to a lesser and those of the $PM_{2.5-10}$ to a greater extent.

The small difference of 4 m between heights of location of the two sampling heads caused considerable differences in the measured concentrations and elemental composition of ambient dust. Such differences may affect interpretation of results of monitoring air for PM_{10} in urban background areas, such as Zabrze. The small difference in heights of head locations may not be significant to measurement of yearly concentrations (long term and/or continuous measurements). However, if an experiment is a measuring campaign to catch short-term phenomena or differences between several areas (in spring/summer), then the type of a sampler and the height of a sampling head location are of great importance. It may apply to majority of urban areas in Poland.

The low contributions of the fine dust and the associated elements to suspended dust in 26.04–03.07.2008 at 2.0 m is not specific of the site but illustrates the effects of resuspension of road and soil dust in summer. In Polish cities, such as Zabrze, unlike in most other European cities, dust and sand are not routinely removed from the streets nor are the streets water-sprayed in summer.

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STĘŻENIA P
M_{2.5,} PM_{2.5,10} I PIERWIASTKÓW ZWIĄZANYCH Z PM NA DWÓCH WYSOKOŚCIACH W OBSZARZE TŁA MIEJSKIEGO W ZABRZU

Celem pracy było zbadanie stężeń i składu pierwiastkowego pyłu drobnego ($PM_{2,5}$) i grubego ($PM_{2,5+10}$) w dwóch punktach wybranych w obszarze tła miejskiego w Zabrzu. Punkty były w zlokalizowane w odległości 20 m od siebie na różnych wysokościach (2 i 6 m). Do pobierania próbek pyłu wykorzystano dwukanałowy pobornik z impaktorem wirtualnym i impaktor kaskadowy. Badania składu pierwiastkowego przeprowadzono wykorzystując spektrometr fluorescencji rentgenowskiej. Badania wykazały heterogeniczność w rozkładzie przestrzennym i składzie pierwiastkowym pyłu zawieszonego w punkcie tła miejskiego. Wniosek ten dotyczy przede wszystkim frakcji cząstek grubych, których, jak wykazano w pracy, imisję w pewnych okresach, na wysokości 2 m, kształtuje wtórna emisja pyłu drogowego, glebowego i inne lokalne "niskopoziomowe" źródła pyłu.