

Anna ZWOŹDZIAK<sup>1</sup>, Izabela SÓWKA<sup>1</sup>, Maria SKRĘTOWICZ<sup>1</sup>,  
Anna WOROBIEC<sup>2</sup>, Alicja NYCH<sup>1</sup>, Jerzy ZWOŹDZIAK<sup>1</sup>  
and Rene VAN GRIEKEN<sup>2</sup>

**PM10, PM2.5 AND PM1.0 INDOOR  
AND OUTDOOR CONCENTRATIONS  
AND CHEMICAL COMPOSITION  
IN SCHOOL ENVIRONMENT**

**STĘŻENIA ORAZ SKŁAD CHEMICZNY PYŁU PM1.0, PM2.5 ORAZ PM10  
W POWIETRZU WEWNĘTRZNYM I ZEWNĘTRZNYM SZKOŁY**

**Abstract:** Simultaneous daily indoor and outdoor measurements of PM1.0, PM2.5, PM10 have been conducted during winter season of 2009/2010 in the secondary school in Wrocław, Poland. Aerosol samples were analysed for mass concentrations and elemental composition. The factor analysis was applied to identify possible emission sources of the PM1.0 fraction. Mean daily PM10 concentrations was 81  $\mu\text{g}/\text{m}^3$  indoors and 54  $\mu\text{g}/\text{m}^3$  outdoors. The corresponding means for PM2.5 and PM1.0 were 62 and 22  $\mu\text{g}/\text{m}^3$  indoors and 46 and 24  $\mu\text{g}/\text{m}^3$  outdoors. There were reported 90 % of days with daily mean exceeding the WHO AQG for PM2.5 – 25  $\mu\text{g}/\text{m}^3$ . In many cases the I/O ratio was higher than 1.0, what means that there are some particles sources inside the school building, particularly for the fractions PM10 and PM2.5. The most abundant elements in the PM1.0 fraction were S, Cl and K. Zn and Pb were the dominant heavy metals. Combustion processes contributed to high concentrations of K, S, As, Cl and vehicular emission to Cu, Pb and Zn.

**Keywords:** PM1.0, PM2.5, PM10 indoor-outdoor particles, elemental composition

Many epidemiological studies allowed to prove the influence of air pollution on human health [1, 2]. The important factors that may affect human health are: the type of air pollutant, its exposure level and personal sensibility (the highest for children and old people). Among others, trace metals associated with aerosol particles with the diameter below 10  $\mu\text{m}$  (PM10), 2.5  $\mu\text{m}$  (PM2.5) and 1  $\mu\text{m}$  (PM1.0) may contribute to particulate toxicity related to an increased risk of respiratory and/or cardiovascular diseases [3, 4].

<sup>1</sup> Faculty of Environmental Engineering, Wrocław University of Technology, ul. Wybrzeże Wyspiańskiego 27, 50-370 Wrocław, Poland, phone: +48 71 320 25 00, fax: +48 71 320 35 99, email: anna.zwozdziak@pwr.wroc.pl

<sup>2</sup> Faculty of Science, University of Antwerp, Universiteitsplein 1, 2610 Antwerp, Belgium, phone: +32 3 265 41 11, fax: +32 3 265 20 04, email: anna.worobiec@ua.ac.be

Much efforts have been made in Europe in improving ambient air quality, but only recently has the international scientific community worried about the indoor air contamination. Indoor levels of many pollutants may be many times higher than outdoor ones [5–7]. Schoolchildren spend as much as 30 percent of their time at school, thus indoor air quality problems in schools are a significant public health concern. Published results regarding *particulate matter* PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1.0</sub> and their chemical composition in school-environment are, at least to the knowledge of the authors, rather scarce in Poland or even they do not exist.

We describe the results of a pilot study evaluating indoor and outdoor PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1.0</sub> concentrations in naturally ventilated school environment in Wrocław. The aerosol samples were analysed for their mass concentrations and elemental composition. The primary purposes of this study were to investigate: (1) differences in the PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1.0</sub> mass concentrations in indoor and outdoor air; (2) elemental composition of PM<sub>1.0</sub> and the *indoor-to-outdoor* ratios (I/O), and (3) the origin of indoor and outdoor PM<sub>1.0</sub> particles.

## Materials and methods

Simultaneous indoor (in the school hall) and outdoor (at the school roof) PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1.0</sub> concentration measurements were conducted in Secondary School No. 13 located in the centre of Wrocław. The measurements were performed on a daily basis (24 h or 2 × 12 h, for one week in December 2009 and in January 2010) with the use of Harvard cascade impactors (MS&T Area Samplers, Air Diagnostics and Engineering, Inc., Harrison, ME, USA). The pumps were set at an airflow of 23 dm<sup>3</sup>/min for PM<sub>1.0</sub>, and 10 dm<sup>3</sup>/min for PM<sub>2.5</sub> and PM<sub>10</sub>. The particles were collected onto 37 mm diameter Teflon membrane filters (PALLFLEX, TK15-G3M). All filters were pre- and post-conditioned in a clean room with environmentally controlled temperature and humidity prior to weighing. Weighing was carried out with an electronic microbalance (Santorius 000 V001).

The bulk elemental composition was analysed with an Epsilon-5 energy dispersive XRF instrument (PANalytical, Almelo, the Netherlands). The details of sampling and the accuracy of analytical methods is given in [8]. In total, 20 elements were determined (Al, Si, S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Se, Sr, Cd, Sb and Pb). Almost all elements were detected in each sample, except for Se, Cd, Al which were not detected in 72, 50 and 52 % of the samples, respectively. Sb was not detected at all.

## Results and discussion

A comparison of mean concentrations with standard deviations and the ranges of indoor particulate matter in 3 sizes including PM<sub>10</sub>, PM<sub>2.5</sub> and PM<sub>1.0</sub> for both sampling campaigns is shown in Fig. 1. PM<sub>10</sub> daily concentrations ranged between 21 and 113 µg/m<sup>3</sup> indoors (mean: 81 µg/m<sup>3</sup>) and between 26 to 87 µg/m<sup>3</sup> outdoors (mean:

54  $\mu\text{g}/\text{m}^3$ ). The respective PM2.5 and PM1.0 daily concentrations varied between 18 and 91, and from 13 to 32  $\mu\text{g}/\text{m}^3$  indoors (means: 62 and 22  $\mu\text{g}/\text{m}^3$ ), and from 24 to 72 and 15 to 41  $\mu\text{g}/\text{m}^3$  outdoors (means: 46 and 24  $\mu\text{g}/\text{m}^3$ ). What is very important that mass concentrations exhibited significant variability. This effect was more pronounced in the PM10 and PM2.5 fractions. 12 h PM10 and PM2.5 indoor concentrations, when available, showed that the values decreased during the night time and on weekends. This indicates that the absence of people inside the school after classes greatly influenced the indoor concentration levels. In the case of PM1.0 concentrations, the situation was not so clear.

The ratio I/O allows to assess whether there are different sources of particulate matter or not. Daily I/O concentration ratios for PM10 varied between 0.8 and 2.5 (mean: 1.5), while for PM2.5 and PM1.0 the I/O ranges were estimated equal to 0.7–1.6 and 0.7 to 1.2, respectively (means: 1.3 and 0.9). In many cases this ratio was higher than 1.0, what means that there are some particle sources inside the school building, particularly for the fractions PM10 and PM2.5. Their existence was confirmed by the correlation analysis. Except for the fraction PM1.0 ( $\text{PM1.0}_{\text{in}} = 0.94 \text{ PM1.0}_{\text{out}}$ ,  $R^2 = 0.87$ ), there were very weak correlations ( $R^2 < 0.5$ ) between concentrations inside and outside the building. This indicates that indoor concentrations of PM10 and PM2.5 were independent on the corresponding outdoor ones. Only the finest fractions could easily penetrate inside the school. Therefore, PM1.0 mass concentrations in indoor and outdoor air and their daily fluctuations were relatively on the same levels.

The mean I/O ratios above 1.0, for both PM10 and PM2.5, were found in many other studies conducted in schools, for instance in London [5], Detroit [6] or Athens [7].

Adverse health effects of particulate matter are mostly attributed to finer particulate matter, thus this fraction was studied more extensively in this work. Table 1 shows indoor mean concentrations of individual elements in the fraction PM1.0 and the *indoor/outdoor* (I/O) concentration ratios. Analysis of the data showed that there were large variations in the range of concentrations observed for each of the elements. The most abundant elements were S, Cl and K. Zn and Pb were the dominant heavy metals. Particularly noteworthy is the presence of significant amounts of Pb, Zn, As, Cu, which can be a serious danger for human health [9].

Much higher indoor concentrations than outdoor ones were observed for crustal elements (Si, Ca, Ti, Fe, Mn). As considered earlier, the reason for this high I/O ratio was probably resuspension of settled fine mineral dust during cleaning (sweeping) and children movement. Many studies suggest that human activity contributes mainly to resuspension of larger particles, that was confirmed in our study for PM10, and to lesser degree for PM2.5, but also the highest I/O ratio values were found for crustal element in the PM1.0 fraction. However, these elements contributed only to about 4 % of the elements mass analyzed in the indoor PM1.0 fraction. Fine particles could remain suspended in air with relatively long time and due to poor ventilation during winter, this process could lead to their accumulation. The I/O concentration ratios for other elements, except for Cl, were at the comparable levels but the values were also above 1.0. Nevertheless, these increases in values for indoor air were small compared with the ones for Si, Ca, Ti and Mn.

Table 1

Indoor mean concentrations of individual elements in the fraction PM1.0 and the indoor/outdoor (I/O) concentration ratios

Element	Mean	Minimum	Maximum	I/O ratio
Si	114.5	17.6	602.4	3.7
K	415.4	193.9	1338	1.1
Ca	33.7	10.0	70.5	6.7
Ti	4.9	0.4	56.9	3.8
Fe	142.4	12.5	1882	2.0
Cr	1.7	< DL*	4.0	1.3
Ni	1.02	0.30	1.63	1.5
Mn	27.8	4.2	319.5	2.2
Cu	23.4	4.2	40.3	1.3
Zn	169.6	50.1	385.5	1.1
As	2.8	< DL	7.4	1.0
Pb	52.4	25.3	99.0	1.1
S	957.3	407.1	1997	1.2
Cl	266.8	43.5	1079	0.3
Cd	0.78	< DL	4.65	1.3

\* < DL – below detection limit.

Simultaneous measurements of PM1.0 mass concentrations at this school showed that the differences between indoor and outdoor air were only within a few percentage range (Fig. 1), and the concentrations were strongly correlated. In the school, no fine

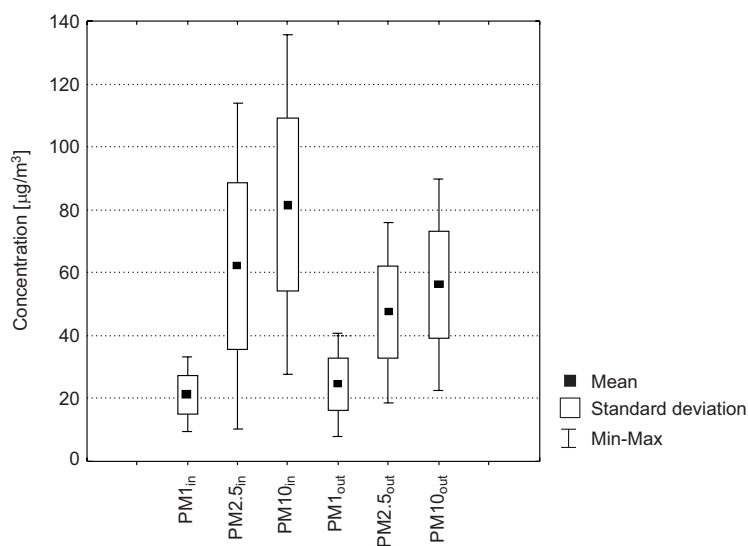


Fig. 1. Mean concentrations, standard deviation and the ranges of indoor (in) and outdoor (out) particulate matter for the fractions PM10, PM2.5 and PM1.0 (cold season, naturally ventilated school)

particle sources such as wood and coal burning and smoking are present. This suggests that the outdoor air is the main contributor to the indoor PM1.0 mass concentrations. As noted earlier some impact of resuspension of mineral dust should be also considered.

In order to estimate the impact of different emission sources on ambient PM1.0 mass concentrations, factor analysis was applied to identify possible emission sources of air particles. Trace elements were used as the markers for the above assessment. Table 2 shows factor loadings from factor analysis for three components. The commonalities for individual elements ranged from 0.76 for As to 0.99 for remaining elements considered. This indicates that three components are quite satisfactory explaining 84 % of the total variance.

Table 2

Factor loadings of individual elements for the PM1.0 fraction (outdoors)

Element	Crustal sources	Coal/wood combustion	Vehicle emission	Commonality
K	0.17	<b>0.84</b>	0.33	0.98
Ca	<b>0.86</b>	0.02	0.08	0.97
Ti	<b>0.99</b>	0.02	0.08	0.99
Fe	<b>0.96</b>	0.13	0.05	0.99
Mn	<b>0.89</b>	0.03	0.41	0.99
Cu	0.06	0.04	<b>0.96</b>	0.97
Zn	0.68	0.36	0.56	0.99
As	0.03	<b>0.81</b>	0.01	0.76
Pb	0.34	0.44	<b>0.80</b>	0.99
S	0.03	<b>0.74</b>	0.29	0.95
Cl	0.08	<b>0.84</b>	0.18	0.85
% variance	44	25	14	84

The first factor that explains 44 % of the variance has high loadings of Si, Ca, Ti, Fe and Mn. It is anticipated that the contribution is from the street dust – soil. However, Ca and Si, which are indicators of crustal matter, were detected at relatively low levels.

Factor 2 includes high factor loadings for K, S, As and Cl that are identified as markers for typical combustion sources. S and As are typical markers for coal combustion in the fine size range [10]. S, K and Cl for wood burning or generally biomass burning [11, 12]. These elements represent above 90 % of the elements mass analyzed in the outdoor PM1.0 fraction.

Factor 3 associated mainly with Cu and Pb (weaker with Zn) usually represents an industrial emission source. However, in our case, in the centre of Wrocław, it is likely that these metals originated from traffic. Studies of some roadway dust particles have shown the presence in small size particles of Pb along with other metals such as Br, Zn and Cu [13]. Cu comes mainly from brake lining wear, Pb from exhaust fumes and Zn from tyre wear and brake lining wear.

## Conclusion

Our results revealed that indoor concentrations of PM<sub>2.5</sub> and PM<sub>10</sub> were higher than outdoor values for the most sampling days. In the presence of high variability of indoor PM<sub>10</sub> concentrations and a weak correlation with PM<sub>1.0</sub> concentrations, the contribution of PM<sub>1.0</sub> in PM<sub>10</sub> varied significantly, from 11 to 59 %. The contribution of PM<sub>2.5</sub> in PM<sub>10</sub> equalled about 78 % for indoor air and 85 % for outdoor air. Daily PM<sub>10</sub> concentrations often exceeded the EU limit values for atmospheric air (50 µg/m<sup>3</sup>).

Concentrations of PM<sub>1.0</sub> in the school hall and in outdoor air were relatively in the same levels. A high correlation between PM<sub>1.0</sub> concentrations inside and outside the building was recorded.

There are some influences on indoor air quality, such as cleaning (sweeping), children movement that cause higher turbulence and stop dust sedimentation. It was proved by higher concentrations of PM<sub>10</sub> and PM<sub>2.5</sub> during the day than the night time and on weekends.

It was observed that a significant amount of heavy metals associated with PM<sub>1.0</sub> fraction, both in indoor and outdoor air, is of anthropogenic origin (combustion processes and vehicular emission). Combustion processes were the most important sources that contributed to high concentrations of As, and vehicular emission to Zn, Pb and Cu.

There is a need to control the particulate matter indoor concentrations. Suitable regulations and standards for indoor air quality should be created, as the finest aerosols can be more dangerous for human's health than the coarse ones.

## Acknowledgement

Presented work was done in the frame of the Project No. N N304 067937: "Identification of air quality impact on the school children's respiratory function".

## References

- [1] WHO (World Health Organization) Europe. Air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide. Global update 2005. Geneva 2006.
- [2] WHO (World Health Organization) Europe. Health risk of PM from long range transboundary air pollution, Copenhagen 2006.
- [3] Timonen, K.L., Pekkanen J., Tiittanen P. and Salonen R.O.: *Occup. Environ. Med.* 2002, **59**, 129–134.
- [4] Moshhammer H., Hutter H.P., Hauck H. and Neuberger M.: *Eur. Respir. J.* 2006, **27**, 1138–1143.
- [5] Wheeler A.J., Williams I., Beaumont R.A. and Hamilton R.S.: *Environ. Monit. Assess.* 2000, **65**, 69–77.
- [6] Yip F.Y., Keeler G.J., Dvonch J.T., Robins T.G., Parker E.A., Israel B.A. and Brakefield-Caldwell W.: *Atmos. Environ.* 2004, **38**, 5227–5236.
- [7] Diapouli E., Chaloulakou A. and Spyrellis N.: *Indoor Built Environ.* 2007, **16**, 55–61.
- [8] Horemans B., Worobiec A., Buczynska A., Van Meel K. and Van Grieken R.: *J. Environ. Monit.* 2008, **10**, 867–876.
- [9] Chapman R.S., Watkinson W.P., Dreher K.L. and Costa D.L.: *Environ. Toxicol. Pharmacol.* 1997, **4**, 331–338.
- [10] Cao L., Tian W., Ni B., Zhang Y. and Wang P.: *Atmos. Environ.* 2002, **36**, 1951–1956.
- [11] Song X.H., Pollissar A.V. and Hopke P.K.: *Atmos. Environ.* 2001, **35**, 5277–5286.
- [12] Cheng Z.L., Lam K.S., Chan L.Y., Wang T. and Cheng K.K.: *Atmos. Environ.* 2000, **34**, 2777–2783.
- [13] Dongarra G., Sabatino G., Triscarib M. and Varricaa D.: *J. Environ. Monit.* 2003, **5**, 766–773.

**STĘŻENIA ORAZ SKŁAD CHEMICZNY PYŁU PM1.0, PM2.5 ORAZ PM10  
W POWIETRZU WEWNĘTRZNYM I ZEWNĘTRZNYM SZKOŁY**<sup>1</sup> Wydział Inżynierii Środowiska, Politechnika Wroclawska<sup>2</sup> Wydział Nauk Przyrodniczych, Uniwersytet w Antwerpii

**Abstrakt:** Jednoczesne pomiary stężeń pyłu PM1.0, PM2.5, PM10 w powietrzu wewnętrznym i zewnętrznym zostały przeprowadzone w szkole średniej we Wrocławiu w sezonie zimowym 2009/2010 roku. Pobrane próbki aerozolu analizowano pod względem wysokości stężenia masowego oraz składu pierwiastkowego. Analizę czynnikową wykorzystano do oceny źródeł pochodzenia cząstek frakcji PM1.0. W powietrzu wewnętrznym średnie dobowe stężenie PM10 wynosiło 81  $\mu\text{g}/\text{m}^3$ , a zewnętrznym 54  $\mu\text{g}/\text{m}^3$ . Odpowiednie średnie dla PM2.5 i PM1.0 wynosiły 62 i 22  $\mu\text{g}/\text{m}^3$  wewnątrz i 46 i 24  $\mu\text{g}/\text{m}^3$  na zewnątrz budynku. Na podstawie przeprowadzonych pomiarów odnotowano 90 % dni ze średnim dobowym stężeniem frakcji PM2.5 powyżej wartości zalecanej w wytycznych Światowej Organizacji Zdrowia, tj. 25  $\mu\text{g}/\text{m}^3$ . W wielu przypadkach stosunek stężeń w powietrzu wewnętrznym (I) do stężeń w powietrzu zewnętrznym (O), tj. I/O, był większy od 1.0, co oznaczało istnienie źródeł pyłów wewnątrz budynku, a dotyczyło do przede wszystkim frakcji PM10 i PM2.5. We frakcji PM1.0 w największych koncentracjach pojawiały się S, Cl i K, a wśród metali ciężkich Zn i Pb. Procesy spalania miały największy udział w stężeniach K, S, As i Cl, natomiast motoryzacja w przypadku stężeń Cu, Pb i Zn.

**Słowa kluczowe:** PM1.0, PM2.5, PM10, powietrze wewnętrzne i zewnętrzne, skład elementarny