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EXPOSURE TO PARTICULATE MATTER AND HEAVY METALS IN THE POPULATIONS OF CENTRAL ZONES OF BELGRADE

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ABSTRACT:

Particulate matter (PM) is hazardous to human health. This paper deals with PM concentrations in ambient and indoor air on three locations (two being in the central parts of the city and another one- near industrial zone of the settlement of Lazarevac, lying in the outskirts of Belgrade). The aim of the paper is to assess the scope of exposure to particulate matter and heavy metals (Pb, NI, Cr, Cd and As) adhered to them. Concentrations of PM in indoor air in the central zones of the city are lower than these measured in ambient air. We concluded that the air is predominant source of heavy metals and metalloids in the environment. Since people spend more than 80% of the time indoors, the control and elimination of local sources of pollution may significantly reduce the scope of exposure to individual pollutants.

Key words: *particulate matter, exposure assessment, toxic metals, indoor air quality*

INTRODUCTION

Particulate matter in the air occurs most frequently as gasses, vapors and solid particles. From the standpoint of environmental health, most important types- solid micro particles, especially these of ≤10μm, have been labeled as suspended particles (PM). Due to their physical characteristics and ability to adsorb numerous pollutants (heavy and toxic metals and metalloids), they have hazardous effects on human health.

The impact of PM on general population has public health significance. This is confirmed by the data from numerous research papers, proving the relation between elevated concentrations (short- and long-term) and health problems that may even have lethal outcome [1]. Health-related problems are varied and depend a great deal upon the concentration and type of the pollutants adsorbed on the particles. With these, PM may act synergistically, additively or cumulatively [2,3].

Exposure to suspended particles, including heavy metals, is especially important both in central and residential zones of the city [4], located near huge industrial complexes. That is why the paper examines the concentration of PM in the two locations in the center of Belgrade (location IPH Belgrade and the kindergarten ("Vrtic"' in 68, Krunska Street) and one location immediately close to the Lazarevac industrial zone - primary school in V.Crljeni. We performed simultaneous measurements in indoor (ID) and open- air (OD) environments, in order to calculate total exposure.

In defining the content of our performance, we hypothesized that the scope of exposure to suspended particles and heavy metals indoors is less prominent than the one measured outdoors (in ambient air). It was also hypothesized that heavy metals adsorbed to PM are predominant in the total exposure.

The aim of the paper is to assess the scope of exposure of the general population to suspended particles and heavy metals. By "general population", we listed adults and the children residing, working or attending preschool and school facilities in the central zones of Belgrade.

METHODS

We used gravimetric analysis of the exposed filters to measure daily (24-hr) concentrations of PM_{10}. As mentioned previously, measuring spots were IPH Belgrade and a school. In addition, using the same method we measured PM_{10} and $PM_{2.5}$ on the location of Kindergarten. We collected samples by low volume sampler Kleinfiltergerat- KGF Sven/ LECKEL LVS3, with inlets for PM_{10} and $PM_{2.5}$. The inlets were 2m away from one another in order to enable independent sampling of different particle fractions. We positioned samplers in the facilities (office, school, preschool facility) and in open- in front of the above facilities. The flow through the sampler was going at 2.3 m^3/h (38 l/min), in accordance with the European Directive EN 12341 and Directive EN 14907 related to the $PM_{2,5}$ sampling procedure.

To collect particles, we used quartz Whatmann 47 mm QM-A filters from the same package. After the gravimetric measurements were over, we cut the filters in smaller parts. The surface of each exposed filter was 13.85 cm^2. The filters were then cut in two surfaces (each 1.76 $cm^{2)}$, to be used for analysis of anions and cations (with total surface of 3.53 cm^2). The remaining 12.09 cm^2 we used for analysis of elements or PAHs.

Gravimetric measurements were performed in the room with controlled temperature (20 ± 10^C) and humidity (50±5%) using Libert Hiross HPM, EMERSON Network Power Model S040A021 VB0200PO Model (Serial No 66097 10001).

Sample digestion was performed according to Standard for Ambient Air Quality (Standard Method for Measurement of Pb, Cd, As and Ni in the PM_{10} fraction of suspended particulate matter CEN/TC 264 N779 [5]. We used microwave oven Anton Paar 3000 for the preparation of samples. The device has a temperature range of 2000^C. Element analysis was performed using ICP/MS Octopole Reaction System, Agilent 7500ce.

In describing the data, we used descriptive statistics- absolute and relative numbers (relations, percent, and proportions), central tendency measures (mean and median values) and variability measures (variation interval and standard deviation).

Data were analyzed by methods of identification of empirical distributions, methods for assessing of the significance of difference and methods for assessing significance of connectedness.

T test for pairs of the data was used to assess significance of the difference in concentrations of the observed pollutants in indoor and ambient air, at each location.

To examine pollutant interrelatedness in indoor and ambient air, we used methods of linear correlation. Interpretation of intensity and direction of connectedness among the variables was done using Pierson correlation coefficient.

In the applied statistical methods, the value of probability of error $p < 0.05$ was statistically significant. In examining correlation, we took into consideration the values of error probability $p < 0.01$, while coefficient value was $r > 0.7$.

Statistical analysis of the data was done using the SPSS for Windows (version 13) statistical package.

RESULTS

For the needs of this Paper, we used the results obtained by analysis of concentrations of PM_{10} and five heavy metals/ metalloids: Pb, Cr, Ni, Cd and As, taken at six micro locations in Belgrade during 2010.

Micro locations:

1. Office within IPHB, Bulevar despota Stefana 54a (Belgrade)
2. Open space within the yard in IPHB (1&2 central part of the city with frequent traffic)
3. Vrtic (kindergarten) in 68, Krunska Street, Belgrade
4. Open space in front of the kindergarten (3&4 central part of the city)
5. Elementary Public School and classrooms in 10, Stevana Filipovica Street (Veliki Crljeni, Lazarevac)
6. Open space in front of the school (5&6 near industrial zone)

We performed measurements in spring (41 days) in Bulevar despota Stefana Street. They lasted 40 days (winter/ spring) in Krunska Street. In Stevana Filipovica Street, measurements lasted 50 days (spring).

Each micro location for indoor air quality was analyzed from the standpoint of construction, technical and sanitary aspect, based on the questionnaire filled by IPHB technician. All three ID spaces are similar from the construction point of view, with soundly isolated windows (PVC or similar).

The results were processed and statistically analyzed. We tested significance of differences and examined connectedness of concentrations of certain pollutants in indoor and ambient air.

Table 1 Concentrations of the examined parameters in indoor air (Location: IPH Belgrade)

Parameter	Mean value	Standard deviation	Minimum value	Maximum value
PM_{10} (µg/m^3)	28.92	15.57	6.90	96.40
As (ng/m^3)	3.00	3.72	<0.001	14.45
Cd (ng/m^3)	0.07	0.18	<0.0001	1.00
Cr (ng/m^3)	1.15	1.82	<0.0005	7.20
Ni (ng/m^3)	1.14	1.87	<0.0001	8.40
Pb (ng/m^3)	13.23	11.81	<0.001	68.40

Table 2 Concentrations of the examined parameters in ambient air (Location: IPH Belgrade)

Parameter	Mean value	Standard deviation	Minimum value	Maximum value
PM_{10} (µg/m^3)	40.92	10.62	15.30	59.90
As (ng/m^3)	3.34	3.53	<0.001	12.48
Cd (ng/m^3)	0.05	0.13	<0.0001	0.70
Cr (ng/m^3)	1.46	2.05	<0.0005	6.00
Ni (ng/m^3)	1.58	1.75	<0.0001	7.10
Pb (ng/m^3)	27.20	20.63	8.40	107.00

We found higher average concentrations of Cd in indoor air within the IPH Belgrade premises. In ambient air, we detected the average values of concentrations for other pollutants.

We found highly significant difference for suspended particles PM_{10} (($t=6.122$; $p=0.000$) and Pb ($t=5.127$; $p=0.000$).

We did not find significant correlation among the concentrations of examined pollutants in indoor air. No significant correlations were established between indoor and ambient environments.

Table 3 Concentrations of the examined parameters in indoor air, PM_{1o},
Kindergarten in 68, Krunska Street, Belgrade

Parameter	Mean value	Standard deviation	Minimum value	Maximum value
PM_{10} (μg/m^3)	38.57	14.18	6.54	76.20
As (ng/m^3)	2.03	1.50	0.61	8.82
Cd (ng/m^3)	0.33	0.22	0.11	1.47
Cr (ng/m^3)	14.41	4.39	8.02	23.89
Ni (ng/m^3)	8.48	3.29	3.63	17.76
Pb (ng/m^3)	13.15	4.84	4.86	26.05

Table 4 Concentrations of the examined parameters in ambient air, PM_{10},
Kindergarten in 68, Krunska Street, Belgrade

Parameter	Mean value	Standard deviation	Minimum value	Maximum value
PM_{10} (μg/m^3)	44.84	16.89	23.40	105.43
As (ng/m^3)	3.22	2.78	0.40	14.00
Cd (ng/m^3)	0.44	0.47	0.10	3.10
Cr (ng/m^3)	14.38	4.49	5.60	23.30
Ni (ng/m^3)	10.56	5.36	3.50	27.00
Pb (ng/m^3)	19.11	8.58	4.30	39.10

Table 5 Concentrations of the examined parameters in indoor air, $PM_{2.5}$,
Kindergarten in 68, Krunska Street, Belgrade

Parameter	Mean value	Standard deviation	Minimum value	Maximum value
$PM_{2.5}$	32.92	10.55	19.00	63.20
As (ng/m^3)	1.89	1.51	0.20	8.90
Cd (ng/m^3)	0.31	0.26	<0.02	1.70
Cr (ng/m^3)	13.14	4.45	5.60	26.60
Ni (ng/m^3)	8.15	3.81	3.50	17.90
Pb (ng/m^3)	11.75	4.25	2.40	23.90

Table 6 Concentrations of the examined parameters in ambient air, $PM_{2.5}$,
Kindergarten in 68, Krunska Str., Belgrade

Parameter	Mean value	Standard deviation	Minimum value	Maximum value
$PM_{2.5}$ (μg/m^3)	40.77	17.64	14.60	97.60
As (ng/m^3)	3.42	3.91	0.70	20.80
Cd (ng/m^3)	0.40	0.42	0.10	2.70
Cr (ng/m^3)	14.03	4.07	7.50	22.10
Ni (ng/m^3)	10.74	4.81	3.00	23.50
Pb (ng/m^3)	18.09	8.59	7.20	54.60

Higher average concentration values of all pollutants were measured in ambient air (around the Kindergarten).

We found statistically significant difference among the examined parameters in ambient and indoor air. The difference related to the following: Cd (t=2.439; p=0.019) and Ni (t=2.685; p=0.011). Statistically high significant difference was found for As (t=4.225; p=0.000) and Pb (t=5.884; p=0.000).

We also found high significant correlation among concentrations of As in indoor and ambient air (r=0.822) and high significant correlation among Cd concentrations in indoor and ambient air (r=0.919).

Table 7 Concentrations of the examined parameters in indoor air, Primary School (Veliki Crljeni)

Parameter	Mean value	Standard deviation	Minimum value	Maximum value
PM_{10} (μg/m^3)	60.33	36.02	14.70	165.50
As (ng/m^3)	2.49	3.42	<0.001	18.00
Cd (ng/m^3)	1.61	3.36	<0.0001	20.90
Cr (ng/m^3)	2.32	3.09	<0.0001	19.40
Ni (ng/m^3)	1.15	3.39	<0.0001	21.60
Pb (ng/m^3)	7.28	4.25	<0.001	16.60

Table 8 Concentrations of the examined parameters in ambient air, Primary School (Veliki Crljeni)

Parameter	Mean value	Standard deviation	Minimum value	Maximum value
PM_{10} (μg/m^3)	52.93	21.23	14.80	119.80
As (ng/m^3)	2.92	2.91	<0.001	9.05
Cd (ng/m^3)	0.21	0.72	<0.0001	4.55
Cr (ng/m^3)	4.42	7.31	<0.0001	34.20
Ni (ng/m^3)	1.65	2.58	<0.0001	12.40
Pb (ng/m^3)	9.12	7.33	<0.001	38.60

In the School inner area, we found higher average values of concentrations for PM_{10} and Cd. In ambient air, we found average values of concentrations for all other pollutants.

We found significant difference in the concentration of Cd (t=2.584; p=0.014) in indoor and ambient air. We did not find any correlation among the concentrations of the examined pollutants in indoor and ambient air.

Table 9 Values of PM_{10} measured in IPH Belgrade and Primary School (Veliki Crljeni)

	IPHB			PS		
	PM_{10}, ID	PM_{10}, OD	PM_{10}, ID/OD	PM_{10}, ID	PM_{10}, OD	PM_{10}, ID/OD
Entire period	28,92± 15,57	40,92± 10,61	0,71	60,32± 30,02	52,93± 21,22	1,14
Workdays	30,14± 17,49	42,16± 10,04	0,71	60,69± 36,67	56,39± 19,72	1,07
Weekends	25,98± 9,45	37,93± 11,78	0,68	59,45± 35,98	44,57± 23,25	1,33

Table 10 PM_{10} and $PM_{2.5}$ ratio in kindergarten, 68, Krunska Str., (in relation to heating season)

	PM_{10}, ID	$PM._{2,5}$, ID	PM_{10}, OD	$PM._{2,5}$ OD	PM_{10}, ID/OD	$PM_{2.5}$, ID/OD	$PM_{2.5/10}$, ID	$PM_{2.5/10}$, OD
Entire period	38,56± 14,18	32,92± 10,54	58,80± 88,41	40,77± 17,63	0,65	0,81	0,85	0,69
Workdays	40,72± 15,25	32,25± 11,13	44,60± 19,61	45,11± 20,18	0,91	0,71	0,79	1,01
Weekends	34,71± 11,54	28,75± 8,19	84,14± 145,12	33,02± 7,52	0,41	0,87	0,83	0,39

To assess the scope of exposure from air, we used the results obtained by mobile devices. These were located within the micro- environments of the observed populations. We also used the data obtained by measurements performed by IPH Belgrade within Local and National networks for the control of air quality.

Data related to concentrations of the pollutants for other environmental media were obtained from the IPH Belgrade. For this occasion, we especially analysed the results from 2010. We took into consideration the 2010 results because IPH had complete examination results of food, water, soil items for general use and other environmental media for that year.

The basic formula used to assess the scope of exposure was the following:

$$\sum \bar{E} = \frac{U_x \cdot C_{a,w,f} \cdot Te}{tt} [\check{z}v]$$

where:

$\Sigma\bar{E}$ – total average exposure
U_x – percentage of intake
$C_{a,w,f}$ – concentration of the examined pollutants in the air, water and foods
Te – length of exposure
tt – body weight
žv – the average life span

Total average exposure ($\Sigma\bar{E}$) is the sum of average exposures to pollutants in the examined environmental media.

Percentage of intake (U_x) is most complex indicator, based on pollutant toxokinetics. It includes all routes of intake and takes into consideration the characteristics of age groups and of their lifestyles [6, 7,8].
Concentrations of the examined pollutants ($C_{a,w,f}$) are these in air, water and foods. However, the concentrations in air are observed as the percentage ratio in certain micro locations.

Working person

- open space (OD) = $2,5^h$ (≈10%)
- car/ any transport vehicle = $1,5^h$ (≈6%)
- indoors (ID) = workplace 8^h (33%)+ residence 12^h (≈50%)

Pre-school/ school child

- open space (OD) = 4^h (≈17%)
- indoors (ID) = kindergarten/ school (33%)+ residence 12^h (50%)

Concentrations of pollutants indoors (residence) were calculated using the values obtained by regular monitoring of ambient air in Belgrade. We took 80% of these values.

Body weight (tt) of adults was 70kg for our research. Children of pre-school age were calculated to be 20kg of weight. Children of school age (lower grades, 1st to 4th) were calculated to be 30kg of weight.

Average life span (l) for females and males was ≈72 years in Serbia.

Although the values of TDI (tolerated daily intake) and ADI (accepted daily intake) are most frequently used in literature, we did not apply them in our research. Instead, we used MRLs (minimal risk levels), issued by ATSDR (Agency for Toxic Substances and Disease Registration), Atlanta 215, 2013 as The Agency takes into account both routes (oral and inhalation) of toxic intake [8].

The formula used to calculate scope of exposure includes the data on quantities of heavy metals with which a person gets into contact, regardless of the manner of this contact.

We established that the concentrations of heavy metals in foods and drinking water are rather low. In most cases, they are even below the MAC level. In case of drinking water (except arsenic), these values are even below the detection threshold [9]. Our results show that the route of entry of heavy metals via foods and drinking water in Belgrade is practically neglectful, in relation to air. These values in the air are not high but due to their specific features (general and continuing exposure), this route of entry must be taken into account. In our case, it is defined as the primary input used to calculate the scope of exposure.

As for the other elements that determine magnitude of exposure defining further the magnitude of the contact, we took the following in order to assess exposure:

- adult pulmonary ventilation = 20 m^3
- child pulmonary ventilation = 5 m^3

In order to assess the so- called inner dose, relevant for this research, we took into account the degree of absorption (through lungs and digestive system) as the major toxic- kinetic feature of heavy metals. Absorption of heavy metals depends on many factors, primarily on the disposal site, physical and chemical characteristics or the particles themselves (solubility, magnitude); one's health and the like. In the concrete case, we took into account the fact that heavy metals enter the human body as previously adsorbed to PM. Therefore, the site of disposal in the body primarily depends on the very kinetics of these particles.

The document "Guidance for Disposal Site Risk", published by Mass Dep' s in 1995 (to be renewed in 2007), cites that 50% of the particles ≥ 10μg are ingested. The remaining 50% reaches the lungs. Therefore,the assessed adsorption of heavy metals [10,6,7,8] is as follows:

- lead (lungs/adults) = 90%
- lead (lungs/children) = 95%
- lead (gastrointestinal tract (GT)/adults) = 10%
- lead (GT/children) = 50%
  ($MRLs_{Pb}$ = not defined because of high sensitivity of humans)
- Ni (lungs/adults) = 30%
- Ni (GT/adults) = 1%
- NI (GT/children) = 5%
  ($MRLs_{Ni}$ = 26 ng/kg/day)
- Cr VI (lungs/adults) = 50%
- Cr VI (GT/adults) = 10%
  (MRLsCr = 86 ng/kg/day)
- Cd (lungs/adults) = 25%
- Cd (GT/adults) = 5%
  ($MRLs_{Cd}$ = 2,9 ng/kg/day)
- As (lungs/adults) = 25%
- As (GT/adults) = 60%
  ($MRLs_{As}$ = 10 ng/kg/day)

In cases where there were not much data about kinetics of heavy metals in children, we used available data for adults, increased by the uncertainty factor [11].

Example of the calculations:
Total exposure to lead ($\Sigma\bar{E}$) of a person working and living in the central zone of Belgrade:

$\Sigma\bar{E} = \bar{E}_a + \bar{E}_w + \bar{E}_f \Rightarrow \Sigma \bar{E}_v$ a=air
w=water
$\Sigma\bar{E}_v = \Sigma\bar{E}_{lungs(pl)} + \Sigma\bar{E}_{gastrointest.(GT)}$ f=foods

$$\bar{E}_{a/pl/Pb} = \frac{U_x \cdot C_v \cdot Te}{70}[\check{z}v]$$

where:
$Ux=90\%$
$Te=4^h+8^h+12^h=24^h$
$C_{a/pl(open\ space)}=5.64\ ng/m^3$
$C_{a/pl(office)}=6.61\ ng/m^3$
$C_{a/pl(house/apartment)}=5.28\ ng/m^3$

$$\bar{E}_{a/pl/Pb} = \frac{90\%\left(4h \cdot 5.64\, ng/m^3 + 8h \cdot 6.61\, ng/m^3 + 12h \cdot 5.28\, ng/m^3\right)}{70} x20$$

$\bar{E}_{a/pl/Pb}=35.74\ ng/kg/day$
$\bar{E}_{a/GT/Pb}=3.96\ ng/kg/day$
$\Sigma\bar{E}_{v/Pb}=39.7\ ng/kg/day$

Table 11. shows the results of calculation of the total exposure to the following metals/metalloids: lead, nickel, chrome, cadmium and arsenic.

Table 11 Exposure to the chosen metals/ metalloids in Belgrade, 2010. Results of the calculations

Metal/Metalloid	Micro location/age group		
	Belgrade (central) adults	Belgrade (central) pre/school children	Belgrade (central) school children
Pb ng/kg/day	39,7	43,4	12,9
Ni ng/kg/day	7,6	9,6	1,1
Cr ng/kg/day	2,45	23,8	8,9
Cd ng/kg/day	0,47	0,39	0,56
As ng/kg/ day	7,4	5,69	11,6

In order to calculate total exposure to PM_{10} (outer dose), we need to add all outer doses in all micro locations at which a person spends certain amount of time:

a) adults (who live and work in the center of Belgrade) = 48,5 $\mu g/m^3$
b) pre-school children (who live and attend kindergarten in the center) = 51,8 $\mu g/m^3$
c) schoolchildren (who live and attend school in V.Crljeni) = 58,5 $\mu g/m^3$
d) limit value LV = 40 $\mu g/m^3$ for PM_{10} and 25 $\mu g/m^3$ for $PM_{2.5}$
e) the WHO recommended value = 20 $\mu g/m^3$ for PM_{10}

DISCUSSION

The analysis of results shows that concentrations of PM_{10} in ambient air were elevated in all three micro locations. They were above LV. The concentrations of PM_{10} in front of the school and inside it were above LV. The concentrations of $PM_{2.5}$ were above LV in the kindergarten and in ambient air.

In the central area of Belgrade, the concentrations of PM_{10} and $PM_{2.5}$ were higher in ambient air than in indoor air. The relationship between the concentrations in ID/OD resembled the data most frequently encountered in the literature [12,13,14].

As for indoor air, the content of PM_{10} and $PM_{2.5}$ exceeded the WHO recommendations (20 µg/m^3) [15] in all three micro locations.

Beside the impact of ambient air and local sources of pollution on air quality in the pre-school facility and school, increased activities of the children during their stay in these facilities also had an impact. It is expected to register elevated reemissions of solid particles in such circumstances, which also contributes to increased concentrations of PM_{10} and $PM_{2.5}$ in indoor air.

We found significant and highly significant differences in concentrations of the majority of examined pollutants in ambient and indoor air.

Elevated concentrations of cadmium in indoor air within IPH Belgrade are most likely the result of proximity of the room with people who had been smoking at that time [16,17].

As for registration of highly significant correlation of the concentrations of As and Cd in indoor and ambient air of the pres-school facility, we may assume that on this micro location we had a significant impact of contaminated ambient air on indoor air, in relation to those two metals.

Calculations of the total exposure to the chosen heavy metals and As showed that pre-school children living in central zones of Belgrade are exposed to elevated concentrations of Pb, Ni and Cr. Schoolchildren living near thermal power plant/heating plant complex (V.Crljeni) are exposed to higher concentrations of As and Cd.

Schoolchildren living and attending school in V.Crljeni are exposed to elevated concentrations of As. These exceed the values defined as levels of minimum risk (MRLs).

CONCLUSIONS

Our research showed that the populations of central parts of Belgrade and these who like near huge industrial complexes are exposed to elevated levels of PM_{10} and $PM_{2.5}$. Numerous pollutants have been adsorbed on the latter particles, including heavy metals and metalloids. We confirmed the hypothesis that the concentrations of PM in indoor air in the central parts of the city are lower that these in ambient air. Another hypothesis we confirmed is that the air is predominant source of heavy metals and metalloids in the environment.

Increased content of PM_{10} and $PM_{2.5}$ in all three micro locations in ambient air that exceeds recommended values might have been expected, since in Belgrade we have been registering (in its central zones) significantly elevated concentrations of these pollutants. The problem exists for many years.

At the same time, our results point to the problems of local sources of ID pollution, and to the problem of overpopulated preschool facilities and schools.

Since people spend approximately 80% of the time indoors, one of the conclusions is that the control and elimination of local sources of pollution in the air can significantly reduce the scope of exposure to certain pollutants.

In view of the fact that in all the samples of air we found presence of heavy metals and metalloids, our conclusion is that the population has a 24-hour exposure to those pollutants – in the working place and kindergarten, at school, during transportation and at home.

Another conclusion is that there is a need to continue with similar research. It would include identification and quantification of the sources of pollution in indoor and ambient air. There is certainly a strong need for Serbia to regulate indoor air quality (office, public facilities, school, kindergarten, hospital, etc.).

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