Environmental Research xxx (xxxx) xxx-xxx



Contents lists available at ScienceDirect

Environmental Research



journal homepage: www.elsevier.com/locate/envres

Environmental and take-home lead exposure in children living in the vicinity of a lead battery smelter in Serbia

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ARTICLE INFO

Keywords: Lead Child Adolescent Statistical models Environmental pollutants Environmental exposure

ABSTRACT

Blood lead levels (BLLs) have been falling steadily worldwide due to restricted use of lead (Pb) and its compounds. although they remain above preindustrial Pb levels. Elevated BLL can still be found in children living near secondary Pb smelters that represent around 50% of Pb production. There have been no studies on Pb exposure in children living in Serbia ever since the 1980s. The aim of this study was to evaluate the BLLs in children living in two villages in Serbia (Zajača, the location of a secondary lead smelter, and Paskovac, 5 km away), identify the primary determinants of children's BLLs, and investigate the impact of BLLs on children's health symptoms and school achievement.

The study was conducted in 2011 on 127 children, aged 1–18 years, whose BLLs were measured using inductively coupled argon plasma mass spectrometry (ICP-MS).

The median BLL in children was $12 \,\mu$ g/dl, with a significantly higher value of $17.5 \,\mu$ g/dl in Zajača, compared to 7.6 μ g/dl in Paskovac. Only 1 out of 75 and 12 out of 52 children from Zajača and Paskovac, respectively, had BLLs below the CDC recommended $5 \,\mu$ g/dl level. Living near the smelter resulted in 19 times, and having a father who works in the plant 4 times higher odds of elevated BLLs. No significant effects of elevated BLLs health symptoms were seen in this study.

BLLs of children living near a battery recycling plant in Serbia, an upper-middle income European country, were in the range and even higher than those of children living in developing countries. For the first time, the contribution of environmental and take-home lead exposure was quantified using mixed-effect modeling, and our results indicate a contribution of 25–40% of the take-home lead exposure to the BLLs of children living in the vicinity of a secondary lead smelter.

1. Introduction

A lead battery is a vital component of 60 million vehicles produced worldwide each year, and around 80% of the world lead production is intended for this use (International Lead Association, 2018). The use of electric bicycles, now representing 20% of lead demand in China, has provided an additional boost to this industry (Chen et al., 2009).

Around 50% of global lead production can be traced back to lead battery recycling, and the auto industry development in Mexico, India, Indonesia and many other Asian countries. Furthermore, the move towards electric vehicles is expected to influence the lead-acid battery market, valued at \$46 billion in 2015, to reach \$84 billion by 2025 (Grand View Research, 2018). Formal secondary lead smelters present occupational hazards but tend to be located in zones approved for this

https://doi.org/10.1016/j.envres.2018.08.031

Received 17 July 2018; Received in revised form 28 August 2018; Accepted 29 August 2018 0013-9351/ © 2018 Elsevier Inc. All rights reserved.

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Fig. 1. Relevant geographical positions of: Panel 1. Serbia; Panel 2. Serbia with neighboring countries, capital city of Belgrade, town of Loznica, and villages of Zajača and Paskovac; Panel 3. Zajača and Paskovac villages; Panel 4. Zajača and the car battery recycling plant (A), school (B), and playground (C).

purpose by the appropriate authority, adhere to local regulation, and control emissions and discharge. On the contrary, informal secondary lead smelters adhere to no regulation, rarely control the pollution they produce, and are often located within residential areas (Carrizales et al., 2006; Taylor et al., 2013). Millions of tons of lead are being released into the environment as a consequence of primitive methods of lead extraction, representing the primary source of particulate lead present in the atmosphere and accounting for more than 78% of lead emission in 2001 (Mao et al., 2008; National Toxicology Program, 2018).

Children below six years of age are particularly susceptible to the toxic effects of lead due to hand-to-mouth activity, better absorption of lead from the gastrointestinal tract, and a neurological system still in development (Lanphear et al., 2002; Lidsky and Schneider, 2003; Ziegler et al., 1978). Lead exposure combined with malnutrition results in increased absorption through the gastrointestinal tract and adds another dimension to this problem in developing countries (Kaiser et al., 2001). Through the 20th century lead exposure was one of the leading causes of morbidity among children and adolescents globally (Canfield et al., 2003; Tong et al., 2000). Chronic low-level exposure to lead has been connected to reductions in school performance, hearing disorders, standardized tests scores (e.g., IQ test), as well as with hyperactive and violent behaviors (Needleman et al., 1996; Nkomo et al.,

2017). It is believed that low-level environmental lead exposure is a largely overlooked risk factor for cardiovascular disease mortality even in developed countries (Lanphear et al., 2018).

The efforts done to control lead release into the environment in the last 20 years, especially the ban of leaded gasoline and paint, have resulted in substantial reductions in children's BLLs around the world. Recent surveys have shown a blood lead level (BLL) of 2.2 µg/dl in the United States and Swedish, and 3.5 µg/dl in United Kingdom's children, although some sub-populations from these countries might still be affected (Cassidy-Bushrow et al., 2017; Roberts et al., 2001). Nevertheless, lead levels remain higher than those which could be contributed only to natural background lead exposure, found in preindustrial humans (Flegal and Smith, 1992; Smith and Flegal, 1992). Lead exposure is still considered an important issue for children in developing countries and additional attention is warranted for children living in lead mining and recycling communities (Fewtrell et al., 2004; Lynch et al., 2000). In developing countries, rural communities are expected to have lower BLLs than urban communities, but this is not the case in populations living in the vicinity of lead mines and smelting facilities, where higher BLLs may occur, particularly among young children (Olympio et al., 2017; Paoliello et al., 2002; Von Schirnding et al., 2003). In addition to the environmental exposure, a significant

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contribution of take-home exposure in children of employees in the battery recycling industries was hypothesized (Gottesfeld and Pokhrel, 2011).

In a recent study, Ericson et al. (2016) estimated there could be up to 30 thousand informal lead-acid battery recyclers in 90 low- and middle-income countries putting up to 16 million people at risk, but this study excluded the Balkan region considering its vicinity to richer European neighbors a protective factor (Ericson et al., 2016). There are almost 2000 papers (500 describing European studies) on environmental exposure to lead in children, but only 2 in the last 15 years dealing with this problem in the any of the six ex-Yugoslavian countries. The most prominent study in the region was the "Yugoslavia prospective lead study", which initiated in the 1980s and ended in early 1990s, following the effects of prenatal, postnatal, and parental lead exposure of infants in Kosovska Mitrovica (the site of a lead smelter) and Priština (a non-exposed population living 30 kilometers away) to early child intelligence, behavior problems, and motor functioning. The authors found small decrements of IQ, modest associations with fine motor and visual motor functioning, and significant but small associations with behavior in exposed children with BLLs of 20-40 µg/dl compared to non-exposed children, with BLLs of 5-10 µg/dl (Wasserman et al., 1998, 1997; Wasserman et al., 2000a, b).

Although current CDC recommendations require monitoring of children with BLLs above $5 \mu g/dl$ (Centers for Disease Control, 2011; Council, 1993), and no safe threshold for BLL in children has been identified (Binns et al., 2007; World Health Organization, 2018), there have been no studies investigating BLLs of Serbian children in the last 30 years. The aim of this study was to determine BLLs in children living near a lead battery smelting plant in Serbia, identify the primary determinants of elevated children's BLLs, and evaluate the influence of these BLLs on children's health symptoms and school achievement.

2. Methods

Children from two villages in Western Serbia, namely Zajača and Paskovac (Fig. 1), participated in this study which was conducted in December 2011. Due to the parents' concerns regarding the health of their children, the Serbian Institute for Occupational Health "Dr. Dragomir Karajović", the Ministry of Health of the Republic of Serbia, and the Loznica Health Center, which is the primary health care service provider in that part of Serbia (see Fig. 1, *Panel 2*), organized this study. The parents were informed about the study objectives and officially accepted to participate in the study by signing the informed consent form.

The study design included a sociodemographic questionnaire about the children's gender, age, place of living, and parents' occupation, and a targeted questionnaire regarding school achievement and the presence of symptoms such as fatigue, heart pounding, forgetfulness, numbness in the extremities, moodiness, and headache. All questionnaire data were self-reported by the participating children and/or reported by their parents. Blood samples were taken from children, and the determination of the blood lead levels was performed. The study was conducted by the principles of the Declaration of Helsinki.

2.1. Setting

Zajača (population: 574) and Paskovac (population: 687) are two villages in Western Serbia, close to the border with Bosnia and Herzegovina (see Fig. 1, *Panel 2*). The lead battery recycling plant is located in the village of Zajača and is around 5 km away from the village of Paskovac (see Fig. 1, *Panels 3* and *4*). Zajača is located at the bottom of a valley and has been the heart of the Serbian antimony industry until the 1980s when, due to the fall in the price of antimony, lead smelting from used lead batteries had started. In the beginning whole lead batteries, including plastic covers, were melted, but due to unpleasant odors and complaints by the inhabitants they began

separating the plastic parts. Initially, workers separated the plastic by smashing the batteries, but the practice was improved between 1994 and 1996 by cutting off the plastic parts using circular saws. This procedure was used until the introduction of a special facility with a closed system in 2012. Although a formal lead battery recycling plant, this plant operated similarly to an informal lead recycling plant as it did not obey local regulations.

The inhabitants of Zajača live in houses which are in close vicinity to the lead smelting plant, with some houses even adjacent to the facility. The elementary school, as well as the children's playground, are located just across the street from the facility (see Fig. 1, **Panel 4**).

2.2. Sampling procedure and laboratory analyses

Blood sampling was done at the Zajača branch of the Loznica Health Center by established guidelines for human biomonitoring of lead exposure (Friberg and Vahter, 1983). Blood was taken from children by venipuncture from the cubital vein into two blood collecting tubes containing sodium heparin (BD Vacutainer Tubes with green BD Hemogard closure). The samples were coded, and then stored and transported refrigerated at 4 °C to the laboratory.

The laboratory of the Louvain Center for Toxicology and Applied Pharmacology, at the Université Catholique de Louvain, Belgium analyzed the blood samples. The exact provenance of blood samples was unknown to the laboratory personnel (blind analysis), and samples were analyzed in a random sequence.

Blood lead levels were quantified using inductively coupled argon plasma mass spectrometry (ICP-MS) on an Agilent 7500cx instrument. Whole blood specimens (500 μ l) were diluted quantitatively (1+9) with a 1-butanol (2%_{w/v}), EDTA (0.05%_{w/v}), Triton X-100 (0.05%_{w/v}), NH₄OH (1%_{w/v}) solution containing Sc, Ge, Rh and Ir as internal standards. The limit of quantification for BLL was 1.0 μ g/dl. Using this ICP-MS validated method, the laboratory has obtained successful results in external quality assessment schemes organized by the Institute for Occupational, Environmental and Social Medicine of the University of Erlangen, Germany (G-EQUAS program).

2.3. Data processing and statistical analysis

Data processing and statistical analyses were performed in the R Language and Environment for Statistical Computing with additional packages (Ihaka and Gentleman, 1996). In tables and text, categorical variables are reported by the number of observations followed by the corresponding percentage of the total. Continuous variables were first plotted, and their distribution was examined both visually as well as tested using the Shapiro-Wilk test for normality. If the condition of normality was satisfied, mean and standard deviation (SD) values are reported in text and tables; statistical analyses were conducted using ttest or one-way analysis of variance (ANOVA) followed by the Fisher's Least Significant Difference (LSD) multiple comparison test. In the case of deviation from the normal distribution, median and interquartile range (IQR) values were reported in text and tables, while non-parametric statistical tests, such as the Mann-Whitney-Wilcoxon and Kruskal-Wallis tests, were used to compare differences between the groups. Associations between children's and parents' characteristics and having an increased BLL (above $5 \mu g/dl$) were obtained by univariate logistic regression, with results expressed as odds ratios (OR) with 95% confidence intervals (95%CI).

Non-Linear Minimization (*nlme*) R package was used to perform the linear mixed-effects analysis of the relationship between characteristics of the children and their BLLs (Pinheiro et al., 2017). All of the characteristics of the children and parents were entered into the model and tested with and without interaction. After the initial analysis (data not shown, but available on request) the age, employment of parents, and the village of residence were entered into the model as fixed effects. Intercepts for subjects were used as random effects. Due to

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Table 1

General characteristics of the children participating in the study and their parents.

	[All] N = 127	Zajača N = 75	Paskovac N = 52	р
Sex				0.549
Male	68 (53.5%)	38 (50.7%)	30 (57.7%)	
Female	59 (46.5%)	37 (49.3%)	22 (42.3%)	
Age (years) ^a	8.50 [6.00; 12.0]	8.00 [5.25; 11.8]	10.0 [6.00; 13.0]	0.278
Age groups				0.975
0–6 years	38 (29.9%)	23 (30.7%)	15 (28.8%)	
7–14 years	70 (55.1%)	41 (54.7%)	29 (55.8%)	
14+ years	19 (15.0%)	11 (14.7%)	8 (15.4%)	
Distance from plant (km)				< 0.001
< 0.3	38 (29.9%)	38 (50.7%)	/	
0.3–1	25 (19.7%)	25 (33.3%)	/	
1+	64 (50.4%)	12 (16.0%)	52 (100%)	
Residence (years) ^a	7.00 [4.00; 11.0]	7.00 [4.00; 11.0]	10.0 [9.50; 10.5]	0.276
Father's age (years) ^a	38.0 [33.0; 41.0]	38.5 [33.0; 41.0]	38.0 [32.5; 42.5]	0.833
Father smelter				0.008
Yes	67 (53.6%)	47 (63.5%)	20 (39.2%)	
No	51 (40.8%)	22 (29.7%)	29 (56.9%)	
Ex-worker	7 (5.60%)	5 (6.76%)	2 (3.92%)	
Father smelter (years) ^a	6.00 [4.00; 15.0]	11.0 [5.00; 17.0]	4.00 [2.12; 5.00]	< 0.001
Mother's age (years) ^a	35.0 [28.0; 37.8]	35.0 [28.0; 37.8]	34.0 [28.0; 37.2]	0.783
Mother smelter				0.011
Yes	9 (7.20%)	9 (12.2%)	0 (0.00%)	
No	115 (92.0%)	64 (86.5%)	51 (100%)	
Ex-worker	1 (0.80%)	1 (1.35%)	0 (0.00%)	

^a Median value and interquartile range.

heteroscedasticity, a model with different variances for each level of the independent variable was used.

3. Results

A total of 127 children participated in this study, of which 52 resided in Paskovac, and 75 in Zajača. The median age was 8.5 years (IQR: 6 – 12), with most of the children (55.1%) being in the age group 7–14. Detailed characteristics of the children who participated in this study and their parents are presented in Table 1. There was no statistically significant difference in the general characteristics between the Zajača and Paskovac children. In Zajača, the median distance of children's homes from the lead smelting plant was 300 m (IQR:

200–980 m), with half of the children (50.7%) residing less than 300 m away from the plant. The median duration of residence in Zajača was 7 years (IQR: 4–11 years), and 10 years in Paskovac.

As far as the parents are concerned, the median age of fathers was 38 years (IQR: 33–41 years), and the median age of mothers was 35 years (IQR: 28–38). More than half of the fathers of Zajača children (67 fathers or 63.5%) and 20 fathers of Paskovac children (39.2%) worked in the smelting plant at the time of the study, with a median work experience of 11 and 4 years, respectively. The difference between Zajača and Paskovac children in the proportion of fathers working in the plant and the median years spent working was statistically significant (p = 0.008 and p < 0.001, respectively). Nine mothers (12.2%) of Zajača children worked at the plant at the time of the study,



Fig. 2. Distribution of blood lead levels in children from Paskovac and Zajača stratified by age groups.

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Table 2

Blood lead levels of children from Zajača and Paskovac aggregated by their general characteristics (median and IQR reported).

		Blood lead level (µg/dl) ^a			
		Zajača N = 75	Paskovac N = 52		
All children		17.50	[12.50; 22.70]	7.60	[5.10; 10.70]
Sex	Male	17.60	[13.00; 22.90]	7.80	[6.22; 10.30]
	Female	16.95	[11.45; 21.52]	6.40	[5.00; 10.70]
Age group (years)	0–6	18.95	[13.95; 23.77]	8.50	[6.90; 11.45]
	7–14	17.60	[14.20; 22.90]	7.65	[5.52; 10.92]
	14+	9.60	[8.92; 12.88]	5.45	[3.82; 6.07]
Distance from plant (km)	< 0.3	17.40	[12.50; 21.70]	/	/
	0.3-1	18.90	[10.25; 23.35]	/	/
	1+	14.45	[14.00; 18.48]	7.60	[5.10; 10.70]
Father smelter	Yes	19.40	[14.55; 23.80]	11.40	[8.60; 12.17]
	No	12.35	[10.17; 17.00]	6.40	[4.40; 7.70]

^a Median value and interquartile range.

Table 3

Association between children's and fathers' characteristics and having the blood lead levels above 5 µg/dl.

	Blood lead level				
	Below $5 \mu g/dl N = 13$	Above $5 \mu g/dl N = 114$	OR [95%CI] ^a	р	p overall
Sex					0.787
Male	6 (46.2%)	62 (54.4%)	Ref.	Ref.	
Female	7 (53.8%)	52 (45.6%)	0.72 [0.21; 2.36]	0.588	
Age ^b	10.0 [8.00; 16.0]	8.00 [6.00; 12.0]	0.90 [0.79; 1.03]	0.117	0.139
Age group					0.037
14+	5 (41.7%)	13 (12.3%)	Ref.	Ref.	
7–14	4 (33.3%)	59 (55.7%)	5.47 [1.24; 26.1]	0.026	
0–6	3 (25.0%)	34 (32.1%)	4.16 [0.86; 24.2]	0.077	
Residence					< 0.001
Paskovac	12 (92.3%)	40 (35.1%)	Ref.	Ref.	
Zajača	1 (7.7%)	74 (64.9%)	19.3 [3.57; 484]	< 0.001	
Father smelter					0.050
No	9 (75.0%)	42 (39.6%)	Ref.	Ref.	
Yes	3 (25.0%)	64 (60.4%)	4.38 [1.20; 21.7]	0.024	
Father's age ^b	40.0 [35.0; 44.0]	38.0 [33.0; 40.8]	0.99 [0.91; 1.08]	0.794	0.536

^a Odds ratio (OR) and 95% confidence interval (95%CI).

^b Median value and interquartile range.

and there was one ex-worker, while none of the mothers from Paskovac ever worked there. This difference was statistically significant (p = 0.011). Due to the small number of cases, fathers who were exworkers, as well as all the mothers were excluded from further analysis.

The median BLL of all children participating in this study was $12 \mu g/dl$ (IQR: 8.0–18.9 $\mu g/dl$). Zajača children had a median BLL of $17.5 \mu g/dl$ while Paskovac children had a median BLL of $7.6 \mu g/dl$, and this difference was statistically significant (p < 0.001). Fig. 2 shows the distribution of BLLs in children from Paskovac and Zajača divided by age group. In Paskovac, most children (73.1%) fell into the 0–10 $\mu g/dl$ BLL category, while in Zajača only 10 children (13.3%) fell into this category. The highest number of children from Zajača (34.7%) fell into the 20–24 $\mu g/dl$ BLL category, followed by the 10–15 $\mu g/dl$ category (28%) and 15–20 $\mu g/dl$ category (24%), and this difference was statistically significant (p < 0.001).

Detailed information on the BLLs between children residing in the two villages, aggregated by the children's and parents' general characteristics is shown in Table 2 (median and IQR values). Boys had a slightly higher median BLL compared to girls, namely 17.6 μ g/dl compared to 16.9 μ g/dl and 7.8 μ g/dl compared to 6.4 μ g/dl in the smelter village Zajača and Paskovac, respectively. In both villages, the age groups 0–6 years had the highest median BLLs, 18.9 μ g/dl and 8.5 μ g/dl in Zajača and Paskovac, respectively. A small drop in the median BLLs was seen in the age group 7–14 years (17.6 μ g/dl compared to 18.9 μ g/dl and 7.65 μ g/dl compared to 8.5 μ g/dl in Zajača and Paskovac, respectively.

Zajača children BLLs in the age group of $14 + (9.6 \,\mu\text{g/dl} \text{ compared to} 18.9 \,\mu\text{g/dl})$. The lowest median BLL of $5.4 \,\mu\text{g/dl}$ was seen in Paskovac 14 + years group of children. In Zajača, there was no significant difference between the median BLL of children living less than 300 m and up to 1 km from the plant, with the median BLLs of $17.4 \,\mu\text{g/dl}$ and $18.9 \,\mu\text{g/dl}$, respectively. Lower BLLs were seen in children living more than 1 km from the plant ($14.4 \,\mu\text{g/dl}$), but still significantly higher than BLLs of $7.6 \,\mu\text{g/dl}$ seen in Paskovac children. Zajača children whose fathers worked in the plant had the highest median BLL of $19.4 \,\mu\text{g/dl}$. Paskovac children whose fathers worked in the plant to the median BLL of $12.3 \,\mu\text{g/dl}$ of Zajača children whose fathers did not work at the plant. Finally, a median BLL of $6.4 \,\mu\text{g/dl}$ was seen in Paskovac children whose fathers did not work in the plant.

Having in mind the recommendation of the CDC, we analyzed the association of gender, age, residence and father's employment in the lead smelting plant with having the BLL above $5 \mu g/dl$ level (see Table 3). Only 1 out of 75 children residing in Zajača had a BLL below $5 \mu g/dl$, and residing in Zajača resulted in 19 times higher odds (OR: 19.3, 95%CI: 3.5 - 484) of having a BLL above $5 \mu g/dl$. Having a father working at the lead smelting plant resulted in more than 4 times higher odds (OR: 4.38, 95%CI: 1.2 - 21.7) of having a BLL above $5 \mu g/dl$. Although children's age as a continuous variable was not a significantly associated with their BLLs, belonging to the 0–6 and 7–14 years age groups resulted in 4.16 (95%CI: 0.86 - 24.2) and 5.47 (95%CI: 1.24 - 26.1) higher odds in having BLL above $5 \mu g/dl$. Children's gender or



Fig. 3. Influence of age, residence (Zajača or Paskovac) and father's employment status on blood lead levels in children.

Table 4

Linear mixed-effects model parameters predicting blood lead levels in children.

Dependent variable: Blood lead lev	el (µg/dl)
Residence	7.719**
Std. error	(0.902)
Father smelter	5.257**
Std. error	(0.914)
Age	- 0.197*
Std. error	(0.077)
Constant	8.196**
Std. error	(0.870)
Observations	118
Log Likelihood	- 350.243
Akaike Inf. Crit.	718.486
Bayesian Inf. Crit.	743.112

Note:

* p < 0.05

**
$$p < 0.01$$

their father's age did not significantly influence these odds.

The linear mixed-effects model analysis (see Fig. 3 and Table 4) indicated that the place of residence (Zajača or Paskovac), father's employment in the lead smelting plant, as well as the child's age significantly predict the BLLs in children. The following equation describes their influences:

$$BLL_{\left(\frac{\mu g}{dt}\right)} = 8.2 + 7.7 \times Residence + 5.2 \times Father smelter -0.2 \times Age$$
(Eq. (1))

where **BLL**_{µg/dl} is the blood lead level, **Residence** is 1 if the child resides in Zajača (0 if in Paskovac), **Father smelter** is 1 if the father works at the lead smelting plant (0 if he does not), and **Age** is the actual age of the child. In practice, this translates to an expected baseline BLL of $8.2 \, \mu g/$ dl (intercept), a 7.7 $\mu g/dl$ increase in BLL for children living in Zajača, and another $5.2 \, \mu g/dl$ increase in children whose father works as a smelter, with a $0.2 \, \mu g/dl$ reduction for each year of the child's age.

Finally, we examined the association between having BLL below or

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Table 5

Association between various symptoms and school achievement and having the BLL above or below 5 µg/dl.

		Blood lead level		
	[All] N = 118	Below $5 \mu g/dl N = 12$	Above $5 \mu g/dl N = 106$	p value
Fatigue				0.359
Yes	15 (12.7%)	0 (0.00%)	15 (14.2%)	
No	103 (87.3%)	12 (100%)	91 (85.8%)	
Palpitations				1.000
Yes	12 (10.2%)	1 (8.33%)	11 (10.4%)	
No	106 (89.8%)	11 (91.7%)	95 (89.6%)	
Forgetfulness				0.595
Yes	9 (7.63%)	0 (0.00%)	9 (8.49%)	
No	109 (92.4%)	12 (100%)	97 (91.5%)	
Numbness of the extremities				1.000
Yes	6 (5.08%)	0 (0.00%)	6 (5.66%)	
No	112 (94.9%)	12 (100%)	100 (94.3%)	
Moodiness				0.691
Yes	21 (17.9%)	1 (8.33%)	20 (19.0%)	
No	96 (82.1%)	11 (91.7%)	85 (81.0%)	
Headache				1.000
Yes	15 (12.8%)	1 (8.33%)	14 (13.3%)	
No	102 (87.2%)	11 (91.7%)	91 (86.7%)	
School achievement				0.352
Good	5 (7.25%)	0 (0.00%)	5 (8.33%)	
Very good	24 (34.8%)	5 (55.6%)	19 (31.7%)	
Excellent	40 (58.0%)	4 (44.4%)	36 (60.0%)	

above the CDC recommendation of 5 µg/dl and various symptoms and school achievement and the results are presented in Table 5. No children in the group of BLL below 5µg/dl reported fatigue, forgetfulness, or numbness of the extremities, while these symptoms were reported by 15 (14.2%), 9 (8.5%), and 6 (5.7%) of children with BLLs above $5 \mu g/$ dl, respectively. Palpitations, moodiness, and headache were reported by one child each in the BLL below $5 \mu g/dl$ group, and by 11 (10.4%), 20 (19.0%), and 14 (13.3%), respectively, in the BLL above 5 µg/dl group. Nevertheless, no statistically significant association was found between reporting these symptoms and having BLL above or below 5 µg/dl. As far as the school achievement is concerned, in Serbia elementary and high school grades range from 1 (not sufficient) to 5 (excellent). In the below 5 µg/dl group, 4 children (44.4%) had excellent, and 5 children (55.6%) had very good school achievement. In the above $5 \mu g/dl$ group, 36 children (60%) had excellent, 19 children (31.7%) had very good, and 5 children (8.33%) had good school achievement. No statistically significant association was found between the BLLs and school achievement in our study.

4. Discussion

Only one out of 75 children living in close vicinity to a lead battery recycling plant in Serbia, an upper-middle income European country, had BLL below the $5 \mu g/dl$ recommendation. In our study, battery recycling operations resulted in elevated median BLLs of $17.5 \mu g/dl$ in Zajača, the village of the lead recycling plant, and $7.6 \mu g/dl$ in Paskovac, the neighboring village 5 km away. Children's age, place of residence, and the employment of the father in the lead recycling plant were the main determinants of BLLs and their contribution was quantified using linear mixed-effect modeling. Nevertheless, no significant effect of elevated BLLs on children's school achievement or symptoms such as fatigue, heart pounding, forgetfulness, numbness in the extremities, moodiness, and headache was found.

In 2012, the CDC proposed the BLL of $5 \mu g/dl$ as a cause for concern requiring monitoring by physicians, lowering the previously existing 10 $\mu g/dl$ recommendation (Centers for Disease Control, 2011). In our study, the most exposed groups of children were the 0–6 years age group living in closest vicinity to the battery recycling plant (BLL 18.9 $\mu g/dl$ in Zajača), and a group having a father working in the plant

at the time of the study (19.4 μ g/dl in Zajača, see Table 2). The median BLL of 12 µg/dl for all the children who participated in this study was above the recommendation mentioned above. Specifically, 74 out of 75 (98%) and 40 out of 52 (77%) children in Zajača and Paskovac, respectively, had a BLL above 5 µg/dl, which would put the significant part of our children (90%) at risk (see Table 3). In 2011 a study of 68 Puerto Rican children aged < 6 years with relatives employed at a battery recycling facility 57% of children had BLLs above 5µg/dl ((CDC), 2012). In our study, children of the same age, as well as across all age groups, had much higher BLLs. Results similar to ours were found in a community-initiated study, conducted 15 years earlier, in Nicaraguan children living near a battery factory, with BLLs 18.8 µg/dl and 16.6 μ g/dl in 6 months – 5 years and 6 years – 13 years old of the exposed children, respectively, and 9.8 $\mu g/dl$ and 6.7 $\mu g/dl$ in the same age of the control neighborhood groups, respectively (Bonilla and Mauss, 1998). In 2011, a review of studies included BLLs of 2284 children residing in proximity to lead battery recycling facilities in developing countries and reported a median BLL of 19µg/dl, a level very similar to the one found in Zajača children. Nevertheless, median BLLs of our children from Zajača and Paskovac were lower than the levels found in the only study on Yugoslavian children, conducted from 1985 until the early 1990s in a lead smelter town Kosovska Mitrovica with controls from Priština. In the mentioned study, BLLs were determined to be $30-40 \,\mu\text{g/dl}$ in the exposed children and up to $10 \,\mu\text{g/dl}$ in the controls (Factor-Litvak et al., 1999).

In general, environmental lead exposure sources for children are mainly the diet, lead in soil and dust, as well as mining and industrial activity (Mielke and Reagan, 1998). Lead-based paint was banned in Yugoslavia in the 1980s and was never commonly used in Serbia, while leaded gasoline was completely removed from use in 2009. It is well known that hand-to-mouth behavior and pica (eating substances not usually eaten, such as soil) also contribute to the blood lead levels in children (Lanphear et al., 2002, 1998). At the time of our study, the Serbian Environmental Protection Agency reported lead levels of 405 mg/kg at a depth of 10 cm and 122 mg/kg at a depth of 30 cm in the soil 150 m away from the plant (near the school). A level of 108 mg/kg at the depth of 10 cm was found in the soil 1.5 km away from the plant, close to Paskovac (Serbian Environmental Protection Agency, 2012). The topsoil (10 cm deep) lead levels in Zajača were 3–4 times

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higher than maximum topsoil lead levels found in the European Union, while the samples close to Paskovac, although lower than the maximum allowed ones, were still 6-7 times higher than the mean levels in the European Union (Tóth et al., 2016). The fact that the school in Zajača, together with the playground, is located across the street from the lead smelting plant (see Fig. 1, Panel 4) should be pointed out, since the playground soil is often a source of lead exposure in children (Etchevers et al., 2015; Mielke et al., 2013). The highest BLLs in younger children, and a reduction of $0.2 \,\mu\text{g/dl}$ for each year increase in age, shown by our model (see Eq. Eq. (1)(Eq. (1)), Fig. 3, and Table 4), can be explained by the reduction of hand-to-mouth behavior and pica, and reduced lead absorption from the gastrointestinal tract in older children. The sharp drop in BLLs of older children from Zajača and Paskovac can be explained by the fact that they usually leave their home, school, and playground at the age of 14 (graduating from elementary school and starting high school) to study in a nearby city of Loznica, more than 10 km away.

Residing in Zajača and having a father who is a smelter resulted in 19.3 and 4.4 times higher odds of having BLLs higher than $5 \mu g/dl$. These odds ratios underline the role of environmental, as well as takehome lead exposure when assessing BLLs in children. The phenomenon of take-home exposure has been reported in pesticide, chrysotile asbestos, and even flour exposure in children of bakers (Fenske et al., 2013; Sahmel et al., 2014; Tagiyeva et al., 2012). Sanders et al. (2014) mention "contact with Pb residues remaining on parents' work clothing" as one of the sources of exposure to lead in children residing in Vietnam battery recycling craft villages (Sanders et al., 2014). In a case report from 20 years ago, the 6-year-old son of a battery reprocessing plant worker was found to have a BLL of above 30 µg/dl, after suffering from chronic constipation, reduced attention span, restlessness, and difficulty functioning at school (Gerson et al., 1996). A meta-analysis of 10 reports found that children of lead-exposed workers had a geometric mean BLL of 9.3 µg/dl with 53% of samples above 10 µg/dl, levels similar to that of Paskovac children, but lower than Zajača children (Roscoe et al., 1999). It is worth noting that the percentage of children from Zajača and Paskovac with excessive lead levels was double of that found in children of New Jersey lead-exposed workers 20 years ago, and several times higher than that found in children with relatives employed at a battery recycling facility in Puerto Rico in 2011 (CDC, 2012; Czachur et al., 1995).

The contribution of the environmental and take-home lead exposure was quantified using a linear mixed-effects model (see Fig. 3 and Table 4). Significant predictors of BLLs were the place of residence, father's occupation (having a father who is a lead smelter), and the age of a child. The expected baseline BLL from our model's parameters was $8.2 \,\mu g/dl$, with additional $7.7 \,\mu g/dl$ added for children residents in Zajača, and 5.2 µg/dl added if the father works at the battery recycling plant. Finally, the child's age reduces the BLL by $0.2\,\mu\text{g/dl}$ for every year of age (see Eq. (Eq. (1) and Table 4). Therefore, a child residing in Zajača, with a father who is a lead smelter, representing the worst case scenario regarding BLL, would have a predicted BLL of around 21 µg/dl, owing 35% to the residence in the vicinity of a lead battery recycling plant, and another 25% to take-home exposure through a parent working in the plant. In children residing further away from the lead smelting plant, the percentage attributable to the take-home exposure is around 40% (e.g., Paskovac children).

The primary prevention of take-home exposure is not to bring work clothes home. At the time of our study, the lead smelting plant in question did not offer cleaning services for the uniforms the workers used, and standard practice was to bring the dirty clothes home and wash them together with other laundry. A pilot study on take-home lead exposure in New Jersey (1995) has shown that none of the children had BLLs above $10 \mu g/dl$ if the parents did not bring home dirty working clothes (Czachur et al., 1995). In a case of increased BLLs among children with relatives employed at a battery recycling facility in Puerto Rico, 85% of vehicle dust samples and 49% of home dust

samples exceeded the U.S. Environmental Protection Agency level of concern of > 40 μ g/ft² (430 μ g/m²). Clean-up of employee homes and vehicles, setting-up shower facilities, shoe washing, and clean changing areas, have resulted in a 9.9 μ g/dl decrease in children's BLL (CDC, 2012). Proper industrial hygiene practices have resulted in BLL decrease of almost 40% in the child of the occupationally lead-exposed worker (Gerson et al., 1996). Reductions of children's BLLs achieved with education, cleaning of clothes and equipment, as well as proper industrial hygiene practices are very similar to the modeled contribution of take-home exposure from our model, which validates our findings.

Reducing lead exposure, especially in children, is a necessary step with important consequences for the society as a whole. High BLLs in children have been associated with substantial reductions in overall school achievement and lower scores on standardized tests (such as the IQ test) (Canfield et al., 2003; Schwartz, 1994). In our study, elevated BLLs were not significantly associated with reduced school achievement nor with various health symptoms examined in the participating children, although fatigue, forgetfulness, numbness of the extremities, palpitations, moodiness, and headache were reported more often in the group with BLLs exceeding $5 \mu g/dl$. As far as school achievement is concerned, most schools in Serbia function by "group comparison" when assigning grades, not necessarily representing an objective (population-valid) evaluation of students' cognitive functioning. Furthermore, school achievement is not sensitive enough compared to a standardized test (IQ test) to reflect the effects of lead exposure at these levels. It was previously thought that the differences in standardized test scores between non-exposed children and children exposed to medium-to-high lead levels were almost undetectable, but when these differences are accumulated in a population, the consequences become more apparent. The cognitive capacity of a person is evaluated through school achievement, which influences the highest attained degree of education and the position one has later in the labor market, which is highly correlated with income and the quality of life. At the society level, reduction of lead exposure has implications on economic activity, tax revenues, as well as the reduction of violence and criminality (Grosse et al., 2002; Olympio et al., 2017). A study has demonstrated that lowering BLLs in Nigerian children by 50% could save up to \$1,000,000,000 per year, while the cost of lead-related health problems has been estimated to \$7,000,000,000 per year (Ogunseitan and Smith, 2007). It is worth noting that the mean BLL found in the Nigerian children was 9.4 µg/dl, similar to that in "unexposed" Paskovac children, and half of that determined in Zajača children. Similarly, a recent report found, among other conclusions, that removing leaded drinking water service lines from homes of children born in 2018 would protect more than 350,000 children and yield \$2.7 billion in future benefits, or about 33% return on investment (Pew Charitable Trusts and Robert Wood Johnson Foundation, 2017).

Our results indicate that a large number of children living in similar conditions, not only in developing countries but also in the middleincome countries of the South East European region, might have elevated BLLs. Identifying formal and informal pollutants, measuring the exposure of children, and conducting detailed industrial hygiene studies of the parents working in different industries could help identify and prevent environmental and take-home lead exposure.

5. Limitations

Both villages in our study are located relatively close to the lead smelting plant and data on BLLs of children living farther away from the plant would give better context to our results and ensure comparison with an "unexposed" population. Unfortunately, BLLs of Serbian children have not been investigated since the eighties, restricting the comparison of these study results to other findings. The participation in our study was voluntary and by the request of the parents, however, the size of our sample compared to the population of the villages (see

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Section 2) guarantees its representativeness. Supplementing the biomonitoring results with a detailed analysis of the lead levels in soil, water and air would shed more light on the sources and media of exposure, but this was out of the scope of this study. A detailed hygienistic analysis of the workers' habits and take-home exposure could enable proposal of more precise and efficient prevention measures. Finally, using a more objective approach to quantifying the possible effects of elevated BLLs on children's cognitive abilities and health would make our results more valid.

6. Conclusion

In Serbia, an upper-middle income European country, BLLs of children residing near a battery recycling plant were in the range of those of children living near lead smelting facilities in developing countries and much higher than those anticipated in children living in developed countries. For the first time, the contribution of environmental and take-home lead exposure was quantified using mixed-effect modeling, and our results indicate a contribution of 25–40% of the take-home lead exposure to the BLLs of children. This approach could prove as being valuable for similar scenarios where children are exposed to combined environmental and take-home pollutants.

Acknowledgments

The research presented in this paper was done as part of the TR 34009, TR 1653014, III 46009, and OI 175036 projects, funded by the Ministry of Education, Science and Technological Development of the Republic of Serbia.

We acknowledge the kind help of Dr. Nina Ćurčić (Geographical Institute "Jovan Cvijić" of the Serbian Academy of Sciences and Arts) in preparing the maps used in this paper.

Competing financial interests declaration

The authors declare they have no actual or potential competing financial interests.

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