



Interdisciplinary Assessment of Children's Lead Exposure in Residential Areas Degraded by Mining (Upper Meža Valley, Slovenia)

Špela Bavec¹ · Teja Čeru¹ · Stanislava Kirinčič² · Matej Ivartnik³ · Viviana Golja² · Janja Turšič⁴ · Klemen Teran¹ · Miloš Miler¹

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Abstract

Children's lead exposure in three mining-impacted residential areas (Črna, Mežica and Žerjav) was modelled using the Integrated Exposure Uptake Biokinetic Model for Lead in Children (IEUBK). Site-specific environmental and dietary Pb source values were determined for modelling. For the first time, dietary exposure from both market and local foods was studied in detail. Children (Group 1: 24–36 and Group 2: 36–48 months) geometric mean blood lead levels (BLLs) were predicted and lead uptake from multiple sources was quantified according to the different dietary exposure scenarios. Biomonitoring data were used for validation. Site-specific soil, house dust and local food Pb contents are higher than legislative and background levels, remaining a cause for concern. Drinking tap water concentrations and outdoor air contents were found in acceptable levels. The determined dietary exposures, ranging from 0.7 to 3.3 µg/kg bw/day, were above the benchmark dose level of 0.5 µg/kg bw/day for developmental neurotoxicity set for Pb in children, indicating a health concern. In general, the estimated BLLs matched reasonably well with the observed BLLs in the Črna and Mežica area for both age groups and in the Žerjav area for Group 2. For Group 1, in the Žerjav area, the output of the IEUBK model overestimated the actual BLLs of the children. For both groups, the primary exposure pathway in Žerjav is from soil/dust, ranging from 55.3 to 84.8%. In Črna and Mežica, soil/dust exposure ranged between 24.2 and 57.8% and between 26.4 and 61.7%, respectively, indicating that dietary exposure predominates when local foods are included. The results of our study also suggest that using the IEUBK default diet value would reduce the dietary exposure up to 25.2% in Črna, 24.2% in Mežica and 8.6% in Žerjav. One of the main findings is that a diet containing local foods can be an important source of lead in mining-contaminated areas.

Keywords IEUBK model · Blood lead level · Soil · House dust · Dietary exposure · Risk assessment

✉ Špela Bavec
spela.bavec@geo-zs.si

Teja Čeru
teja.ceru@geo-zs.si

Stanislava Kirinčič
stanka.kirincic@nijz.si

Matej Ivartnik
matej.ivartnik@nijz.si

Viviana Golja
viviana.golja@nijz.si

Janja Turšič
janja.tursic@gov.si

Klemen Teran
klemen.teran@geo-zs.si

Miloš Miler
milos.miler@geo-zs.si

¹ Geological Survey of Slovenia, Dimičeva ulica 14,
1000 Ljubljana, Slovenia

² Centre for Environmental Health, National Institute of Public
Health, Trubarjeva cesta 2, 1000 Ljubljana, Slovenia

³ Regional Unit Ravne na Koroškem, National Institute
of Public Health, 2390 Ravne na Koroškem, Slovenia

⁴ Environmental Agency of the Republic of Slovenia, Vojkova
1b, 1000 Ljubljana, Slovenia

Introduction

The recommended blood lead level (BBLs) for children was lowered from 50 to 35 $\mu\text{g/l}$ in year 2021 and it is represented as a “reference value”. However, there is no safe level of Pb for children or adults (US CDC, 2024). Lead-induced health effects are especially worrisome for preschoolers, since their developing nervous, immune, digestive and other bodily systems are easily affected (Grandjean and Landrigan 2006; Collin et al. 2022; Samuel et al. 2022), and exposure to potentially toxic elements (PTEs) and other neurotoxic chemicals can have an impact on various diseases later in life (Roberts et al. 2009). Children have a higher absorption rate, lower lead excretion and increased vulnerability of their central nervous system (immature lead-targeted organs and systems) (Cleveland et al. 2008). Studies have shown that preschoolers are more vulnerable to incidental or intentional dust and soil particles ingestion and inhalation during indoor (floors/carpets) and outdoor (yards and playgrounds) playing activities (Roberts et al. 2009; Bradham et al. 2013; Özkaynak et al. 2022) than other age groups of children. Overall mean soil and dust ingestion rate for toddlers and young children (6 m–11 yr) is about twice that of infants (0–6 m) and three times that of teenagers and late adolescents (Özkaynak et al. 2022). House dust and soil are one of the main sources of children’s exposure to lead (Rudel et al. 2003, 2008; Liroy 2006; Mielke et al. 2011; Taylor et al. 2013). Children are also exposed to lead through ingestion of food and water, especially in some underdeveloped countries, where the cooking water contains a relatively high concentration of Pb (Akers et al. 2020). In some cases, lead contents in food are increased and may even exceed food safety guidelines (Li et al. 2016). Children can also be exposed to lead by inhaling lead dust from lead-based paint or by playing with toys containing lead-based paint (Njati et al. 2019), but these influences play a very minor role in environmentally contaminated sites. To assess children’s health risk from lead exposure, the United States Environmental Protection Agency (US EPA) developed the Integrated Exposure Uptake Bio-Kinetic (IEUBK) model. The model has been widely used in human health risk assessments, primarily at contaminated mining and smelting sites (Carrizales et al. 2006; Ivartnik and Eržen 2010; Jež and Leštan 2015; Li et al. 2016; Zhang et al. 2017; Heuskinveld 2021; Brown et al. 2023), but also in the urban environments (Gulson et al. 2018; Zhong et al. 2017).

Due to the 300-years history of lead–zinc mining, smelting and also lead recycling activities, the Upper Meža Valley (Slovenia), which has a population of about 7.500 inhabitants, is heavily polluted with Pb, Zn and

Cd (Svete et al. 2001; Vreča et al. 2001; Šajn 2006; Fux and Gosar 2007; Gosar and Miler 2011; Miler and Gosar 2013, 2015, 2019; Finžgar et al. 2014; Gošar et al. 2015; Miler et al. 2022; Goltnik et al. 2022). Investigations revealed that, in addition to mineralized rocks, past Pb-ore processing emissions, mining waste and the Pb-waste recycling industry are major sources of enriched metal contents in soil, sediment, attic, road and household dust (Šajn et al. 2000; Miler and Gosar 2019; Teran et al. 2020; Miler et al. 2022; Pučko et al. 2024). In order to estimate the population’s lead exposure, biomonitoring of blood lead level (BLL) in children has been carried out since 2004 (Ivartnik et al. 2021). The Upper Meža Valley was proclaimed as a brownfield site and received a *Program of Measures to Improve the Quality of the Environment* (Official Gazette No. 119/07 and 44/22—ZVO-2; ZVO is an abbreviation for the national Environmental Protection Act), hereafter referred to as the Program of Measures. Implemented between 2007 and 2022, the program aimed to protect human health, especially that of children. One of the main objectives was to reduce Pb levels in children’s blood (95% of children below 100 $\mu\text{g/L}$). The Program of Measures included the replacement of contaminated soils, asphaltting of unpaved roads, wet cleaning of paved public surfaces, roofs and facades, establishing vegetation covers on bare public surfaces, particularly those intended for children’s activities, etc. In addition, safe gardening locations have been arranged, food subsidies have been provided in public institutions (schools, kindergartens) to ensure easy access to healthy food, and regular monitoring of air, soil and water has been set up. A working group has been established to raise awareness of Pb exposure and possible health effects.

The lead exposure of children improved rapidly in the early years of implementing the measures, but no further improvement was observed after 2010 (Ivartnik et al. 2021). The first children’s exposure modelling using IEUBK was based on site-specific concentrations of soil, water and dust, measured between 1989 and 2001 (prior Program of Measures) and validated by biomonitoring results up to 2007. The results showed that more than 80% of the total lead intake occurred through soil and dust (Ivartnik and Eržen 2010). Later, systematic soil sampling, investigation and remediation were carried out. The results showed that with EDTA, soil washing reduced soil Pb concentrations by up to 67% and decreased the concentrations of in vitro bioaccessible Pb in the simulated human gastric phase by up to 3.2 times (Jež and Leštan 2015). The IEUBK model predicted that, after soil remediation, the number of locations where the expected blood Pb level in children exceeded the threshold of 100 $\mu\text{g/L}$ would decrease by up to 91%. However, both studies (Ivartnik and Eržen 2010; Jež and Leštan 2015) emphasized that dietary exposure was not studied in detail,

although the consumption of home-grown vegetables in the Upper Meža Valley is a common practice.

To our knowledge, only in a few studies, conducted in areas affected by mining and mineral processing, the IEUBK default diet values were replaced with national dietary data. Zheng et al. (2013) used national dietary values from FSANZ (Food Standards Australia New Zealand 2003) in Australia. However, they did not consider local dietary values but emphasized in their conclusion the need to include this aspect in future work for a better understanding of lead exposure. In China, Li et al. (2016) found that dietary exposure could be the major source of lead intake (median of 83.39%) for children living in mining areas. They used data from the 2002 Chinese National Nutrition and Health Survey's food categories to estimate dietary lead intake, as food samples were not collected from local farmlands. Zhang et al. (2017) determined Pb intakes from local rice, vegetables, pork and chicken, but found out that diet was the second most significant exposure pathway, contributing an average of 8.86% of total lead. This is because home-grown foods accounted for about 17% of the whole diet due to the limited farmland in this region. It seems that in areas that were heavily polluted by historical Pb mining and processing activities, the dietary lead intake aspect was not given much attention due to the high levels of Pb in soil and dust. This study aims to address this knowledge gap, with one of its key questions focussing on how local food consumption influences overall Pb exposure in a highly contaminated area like the Upper Meža Valley. This question becomes even more important when considering the habits of the local population in the study area, most of whom grow vegetables in their own gardens. Therefore, in this study, the extensive environmental (soil, house dust, air, drinking water), dietary (food, food contact ceramics) and biomonitoring (blood) data from the Upper Meža Valley were gathered, processed and interpreted by an interdisciplinary scientific working group. The main objectives were (1) to establish site-specific levels of soil, outdoor air, indoor dust and drinking water, reflecting environmental conditions after the implementation of Program of Measures; (2) to determine dietary exposure levels by analysing both market-sourced and locally produced foods, including contributions from ceramic food contact materials; (3) to predict and validate blood lead levels (BLLs) in children aged 24–48 months according to the different dietary exposure scenarios; and (4) to quantify the contribution of different lead sources to identify the major exposure pathways.

This study was also conducted as a case study to support the efforts of the Partnership for the Assessment of Risks from Chemicals (PARC) (PARC 2024) in advancing next-generation risk assessment (NGRA). NGRA aims to aggregate multiple sources and routes encompassing both living and working environments, as well as indoor and outdoor

settings. The authors of this study are members of the PARC scientific community, which is committed to enhancing the understanding of the mechanism of action of chemicals and their role as upstream determinants of diseases. This knowledge is crucial for mitigating the effects of exposure to hazardous chemicals on environmental and human health.

Methods

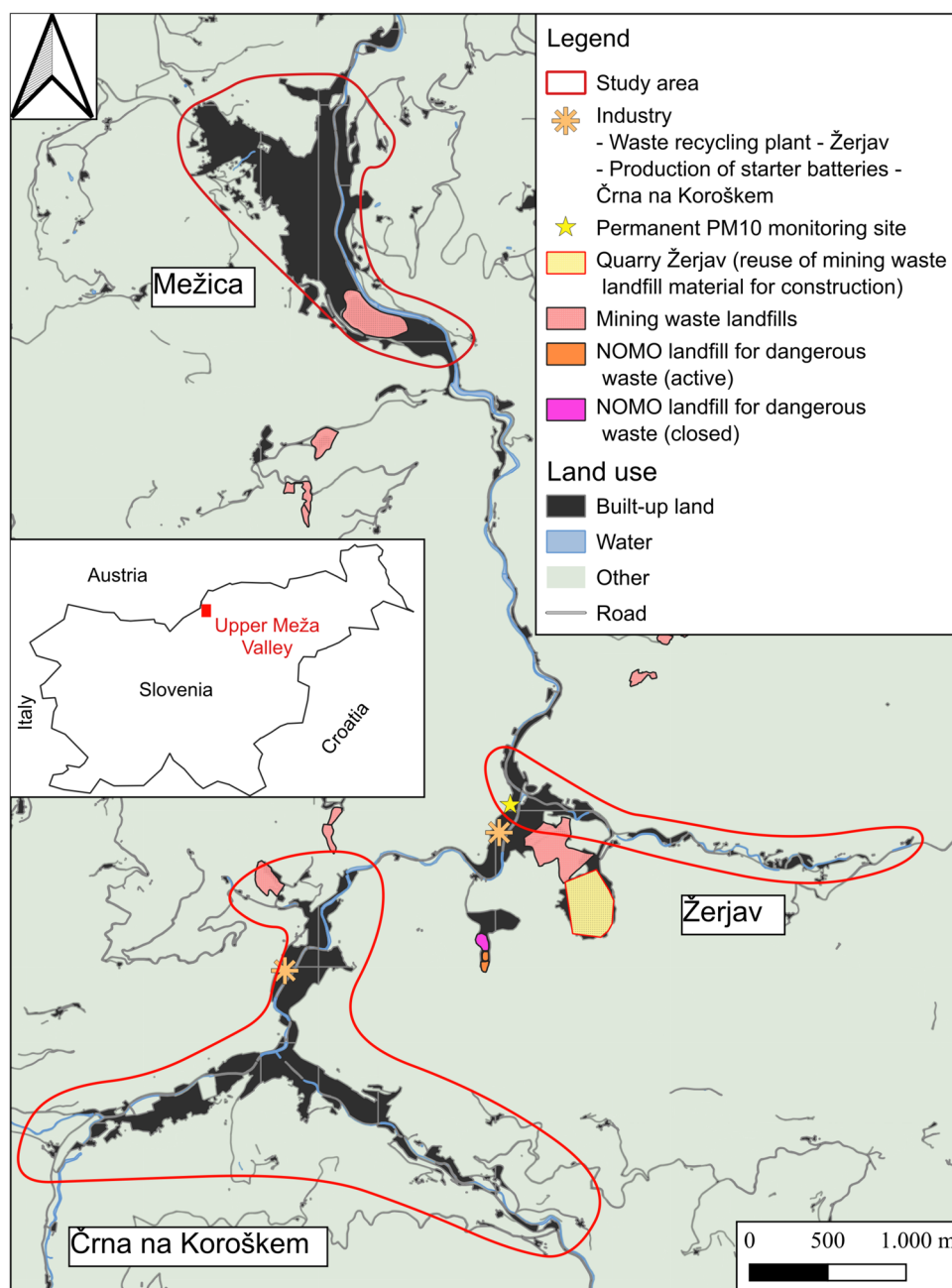
Study Site and Study Population

The study site is presented in Fig. 1. The environment in the Upper Meža Valley (Slovenia) is heavily burdened with potentially toxic elements (PTEs) due to historical Pb, Zn and to a lesser extent Mo mining and smelting activities. Three residential settlements (Mežica, Žerjav and Črna na Koroškem (hereafter referred to as Črna) (Fig. 1) were selected as case study areas. According to the Statistical Office of the Republic of Slovenia (SiStat database), the population in 2023 was 2131 in Črna, 362 in Žerjav and 3127 in Mežica. The annual number of children aged 0–4 years living in Mežica between 2008 and 2023 varied between 111 and 150, in Črna between 76 and 104 and in Žerjav between 7 and 13. In addition to immissions in soil, dust, air and other environmental media (Šajn and Gosar 2004; Gosar and Miler 2011; Miler and Gosar 2013, 2019; Gošar et al. 2015; Goltnik et al. 2022), major sources of Pb include the mining waste landfills located across the valley (Gosar et al. 2020; Miler et al. 2022), industrial activities and the Žerjav quarry (Fig. 1). At the Žerjav quarry, mining waste is recycled into construction materials, which are applied in the local and also more distant environments (Zupančič et al. 2021). Pb-based waste recycling also takes place in Žerjav, contributing to local contamination (Miler and Gosar 2013; 2019), with residual waste disposed of at a nearby hazardous waste landfill (NOMO). The production of starter batteries is carried out in Črna.

Modelling and Collection of Site-Specific Data

The U.S. EPA's Integrated Exposure Uptake Biokinetic (IEUBKwin v2.0 Build 1.72) software model was employed to assess lead exposure from soil, dust, drinking water, air and diet. The history of model development is provided in the User's Guide (US EPA 2021). Following the User Manual's recommendation, site-specific data were used instead of default intake values. The IEUBK software was run in a single simulation mode for each scenario. Based on the Pb uptakes results from these simulations, the contribution of each source to the overall predicted BLLs was calculated. Modelling was focussed on two age groups (Group 1 = 24–36 and Group 2 = 36–48 months). The

Fig. 1 Study areas and important sources of lead excluding immissions in environmental media (map base data sources: mining waste (Gosar et al. 2020); Žerjav quarry (Mining Registry Book 2023); NOMO (OVD No. 35406-12/2020-26); industry (EAS 2024); land use (LU DB 2024))



predicted BLLs were validated against the observed BLLs. Additionally, the predicted BLLs were also calculated for the general population of Slovenian children and compared with those of children from the Upper Meža Valley.

Upper Meža Valley

Spatial data on Pb in soil, indoor dust (household dust from vacuum cleaner bags) and drinking tap water were obtained from national databases. Data were sourced from the Slovenian Environment Agency (ARSO) (soil), Geological survey of Slovenia (GeoZS) (dust) and public water distribution

system records (tap water). The free QGIS 3.28.2 software was used to extract and process the spatial data. The case study areas were defined with red polygons (Fig. 1) around the residential areas, where available environmental data were concentrated. The intersection tool was used to extract point data representing sampling locations with associated Pb concentrations. Individual point data outside the defined residential polygons, representing rural outskirts, were scarce (mostly soil) and critically lacked paired measurements for house dust, PM10 and local food Pb concentrations, which are essential inputs for the IEUBK model. Therefore, to ensure a complete exposure assessment

for each included location, these isolated data points were excluded from the case study datasets. Case study datasets were prepared for further analysis using Microsoft Excel software. The *soil* dataset comprised 35 lawn topsoil samples (0–5 cm) and 61 garden (including fields) soil samples (0–20 cm) (unpublished). Soil was sampled as a part of a soil monitoring program (between 2008 and 2022). Each year, random and targeted (e.g. at households of children with elevated BLLs) sampling locations were established around residences, where children participating in the biomonitoring program, lived. In the case of multiple sampling at the same site, the arithmetic mean of Pb content was calculated to represent that location. The *household dust* dataset comprised 18 samples collected during different research surveys; Two samples in Žerjav in 2011 (Miler and Gosar 2019; analytical fraction < 125 µm) and 1 sample in Žerjav, 2 in Črna and 2 in Mežica in 2016 (Teran et al. 2020; analytical fraction < 63 µm). The remaining samples (6 in Mežica, 1 in Žerjav and 4 in Črna; analytical fraction < 125 µm) were collected in 2023 from households where children who participated in the biomonitoring program (2023) resided (unpublished data). The *tap drinking water* dataset included 12 samples (unpublished) from 3 monitoring stations (one in each study area). The tap water was sampled according to SIST ISO 5667-5:2007 in the period between 2021 and 2023. Due to the sensitive nature of the data, which could potentially reveal the identities of participating children or a resident, the sample locations of soil, house dust, BLLs and tap water were not displayed on the map. The Pb contents in *outdoor air* (particulate matter with a diameter of less than 10 microns—PM10) were determined using archived monitoring data (between 2009 and 2022) from measurements conducted by ARSO at the permanent monitoring station in Žerjav (Fig. 1). As the station is located near an industrial site with unique weather conditions, where extreme Pb contents were recorded, factors (Table 4) were applied to estimate the representative outdoor air Pb PM10 levels in the case study areas. These factors were derived from PM10 Pb measurements, which were carried out during special sampling campaigns, including one-year measurement period (autumn 2007 to autumn 2008) in Žerjav, Črna and Mežica and additional measurements in autumn 2018 in Črna and at various locations in Žerjav), when simulation measurements took place (Koleša et al. 2008; Ivartnik et al. 2019).

Overall chronic *dietary Pb exposure* values of average toddler's consumers were estimated deterministically (FAO/WHO 2009; EFSA 2011). The lower bound (LB), middle bound (MB) and upper bound (UB) dietary exposures to Pb in a single food category were calculated by multiplying the arithmetic mean consumption values by the LB, MB and UB arithmetic mean Pb concentration in food, respectively. Left-censored concentration results (non-detects) were substituted with zero, LOQ/2 and LOQ, to derive the LB, MB

and UB arithmetic mean Pb concentrations, respectively (EFSA 2010a). The LB, MB and UB arithmetic mean Pb concentrations for different food groups were obtained from the Slovenian market (Kirinčič et al. 2019), from local foods as well (Ribarič-Lasnik et al. 2002; Kirinčič et al. 2017) and missing Pb concentrations taken from the EFSA database (EFSA 2012). These values are presented in Table S1. Consumption data for toddlers (1–3 years) were sourced from EFSA summary statistics for Slovenia (EFSA 2024), which is based on the national diet study (SI-MENU 2018), and are provided in Table S2. Local foodstuff included were leafy vegetables, root vegetables, potatoes, tomato and peppers, parsley—aromatic herb, cattle milk, hen eggs and brook trout eggs (wild) (Table S1). The overall chronic dietary exposure for the average consumer was calculated as the sum of exposure increments from all food groups. Due to the lack of data on local garden produce consumption, three dietary exposure scenarios were developed for children for use in the IEUBK models. Based on consuming market versus local food consumption (Table 5), Scenario D1 represents children consuming only Slovenian market foods (general reference scenario). Scenario D2 assumes most consumed food is from the market, except for half of the intake from specified local foods. Scenario D3 involves children consuming specified local foods instead of market foods, with the remainder sourced from the market.

Ceramic food contact materials (423 pieces of ceramic holloware including various types of cups, containers and deep plates) were selected for determination of exposure as they are the most significant source of lead among food contact materials. The migration of lead was monitored between 2013 and 2023 (unpublished data) within the framework of official control of the Slovenian market. The samples were taken by the Health Inspectorate of the Republic of Slovenia (ZIRS). The migration tests giving concentration of lead in simulant 4% acetic acid were performed under the test conditions described in the Directive 84/500/EEC (Council Directive 1984): four replicates of each sample were filled with 4% acetic acid for 24 ± 0.5 h at the temperature of 22 ± 2 °C.

The first step of the exposure assessment was performed using Eq. (1):

$$\text{Estimated exposure to lead} = (\text{Concentration} \times \text{Food consumption}) \quad (1)$$

Food consumption data for fruit and vegetable juices and nectars (used as a worst-case scenario) for toddlers were taken from the Slovenian national food consumption survey (SI MENU 2018), published in the EFSA Comprehensive European Food Consumption Database (EFSA 2024).

For the second step of the exposure assessment, allocation, substitution and decreasing factor were considered for three different scenarios for food contact materials (Table 6).

An allocation of 0.85 for holloware was assumed because it is proportional to percentage of holloware samples collected during the period of 2013–2023. To reduce the overestimation, arising from the assumption that ceramic articles constantly release metals at the level of the first migration test over repeated exposure, average decreasing factor of 20% was applied (Beldi et al. 2016). Data below the limit of quantification were censored and replaced by zero (lower bound, LB), by half of the LOQ (middle bound, MB) and by the LOQ (upper bound, UB).

Blood lead level (BLL) data of 3-year-old (24–48 months old) children was obtained from the National Institute of Public Health (NIJZ) database to validate the IEUBK's model predicted levels. Biomonitoring of children's BLLs has been conducted annually since 2004. Access to blood Pb data (up to 2022), including spatial coordinates and age, was approved following an ethics review by the NIJZ. Given the lead exposure remained relatively constant with minor fluctuations after 2010 (Ivartnik et al. 2021), the BLL measurements from 2011 onwards were extracted from the database using the same methodology as for the environmental data. The biomonitoring data were divided in two groups (Group 1: 24–36 months and Group 2: 36–48 months). Boxplot distributions of BLLs across the studied areas were plotted using R version 4.3.1 and RStudio software (utilizing packages by Wickham (2016) and Wickham et al. (2023)) and compared with the predicted BLLs.

Slovenian Territory Data

The site-specific data for Slovenian territory were obtained from existing studies. Soil and household dust values (Table 1) were taken from Teran (2020), providing the most recent (sampled in 2016) Pb content measurements. Additionally, the samples were homogeneously distributed across urban and rural areas, with the most extensive coverage for

household dust sampling. Topsoil samples (0–5 cm) were prioritized as the most relevant layer for exposure assessment. A lower-bound level of 0.5 µg Pb/L (Kirinčič et al. 2019) was used for drinking water (tap water), because in the majority (74%) of samples, the Pb concentration was < LOQ. The PM10 Pb contents (expressed as arithmetic means) were calculated from the average annual Pb contents reported for PM10 monitoring stations in urban areas (Celje, Ljubljana and Maribor) and a rural area (Iskrba) (Table 4) between 2008 and 2021 (Koleša 2023). For dietary Pb exposure, the lower bound (LB) estimate of the average Slovenian toddler (aged 1–3 year) was used, assuming consumption of only market-available foods (as determined in this study). Human biomonitoring (HBM) data for Slovenian children aged 24–48 months are not available.

Chemical Analysis and Quality Control

All environmental chemical analyses of the samples from the Upper Meža Valley were conducted in accredited laboratories according to ISO/IEC 17025. Soil sample Pb contents were measured using inductively coupled plasma mass spectrometry (ICP-MS) after aqua regia digestion (1:3), in accordance with SIST EN16171:2017 and the Pb concentrations in tap water were determined according to ISO 17294-2: 2016 at the National laboratory of Health, Environment and Food, accredited by the Slovenian Accreditation under accreditation number LP-014. The PM10 Pb contents on filters were analysed following EN 14902:2005 (microwave digestion using HNO₃ and H₂O₂ followed by ICP/MS analysis) at the Chemical analytical laboratory of the Slovenian Environment Agency, accredited by Slovenian Accreditation under accreditation number LP-030. The Pb contents in house dust samples were measured by ICP-MS after a modified aqua regia digestion (1:1:1 HNO₃:HCl:H₂O) at Bureau Veritas Minerals, Mining laboratory services Canada.

Table 1 Basic statistical data of Pb contents (mg/kg) in topsoil and house dust of Slovenian territory (refers to general/non-mining areas, while “Upper Meža Valley” data are site-specific)

Source		Soil			Household dust			
Reference		Gosar et al. (2019)	Teran (2020)		Šajn (1999)	Teran (2020)		
Depth/fraction		0–10 cm	0–5 cm		< 0.125 mm	< 0.063 mm		
Sample collection		1990–1993	2016		no data	2016		
Parameter	Unit	Slovenia	Slovenia rural area	Slovenia urban area	Slovenia rural area	Slovenia urban area	Slovenia rural area	Slovenia urban area
Count		819	105	143	41	23	101	137
Minimum	mg/kg	6.2	14	18	62	152	15	7.9
Maximum	mg/kg	850	1700	1300	523	516	2500	890
Arithmetic mean	mg/kg	40	/	/	146	254	/	/
Median	mg/kg	34	36	51	138	236	54	61
Geometric mean	mg/kg	34	/	/	/	/	/	/

Local food samples were analysed at accredited laboratories: ERICo Velenje (Institute for Ecological Research, now Eurofins) and the Centre for Pedology and Environmental Protection at University of Ljubljana's Biotechnical faculty. Methods are detailed in Ribarič-Lasnik et al. (2002) and Kirinčič et al. (2017), respectively. For Slovenian territory, the methodology and quality control for Pb measurements in soil, house dust, PM10 and market food samples are described in referenced studies (Kirinčič et al. 2019; Teran et al. 2020; Koleča 2023).

Whole blood lead levels (BLLs) were analysed at the Clinical Institute of Clinical Chemistry and Biochemistry, University Medical Centre Ljubljana. From 2011 to 2013, electrothermal atomic absorption spectrometry (ETAAS) was used, while inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7700, Japan) has been employed since 2014. The laboratory successfully participated in the INSTAND External Quality Assessment Scheme for over 8 years, using validated ICP-MS methods.

Results and Discussion

Environmental Media

The basic statistical data of the Pb content in soil are presented in Table 2. In general, lead content of about half of the lawn and garden soil locations in all studied areas exceed the critical immission value for soils of 530 mg/kg (Official Gazette No. 68/96, 41/04—ZVO-1 and 44/22—ZVO-2). The critical levels represent the Pb contents in soil, at which the soil is not suitable for growing crops used for human consumption or animal feed, as well as for filtering and containment of water. Compared to previous studies (Table 2), the median Pb contents in soil from this study are lower (about

two times lower in Črna and Mežica and about six times in Žerjav). This could be mainly a consequence of the different sampling strategies across studies, as well as the inclusion of soil samples from more frequently visited children's playgrounds and some gardens, where remediation was conducted under the Program of Measures. Nevertheless, soil pollution remains a significant issue, particularly in garden soils. Many residents own small gardens and grow their own vegetables, which is a common practice in the area. It has been shown that the average Pb concentrations, as well as the proportion of non-compliant samples (EU limit values from Regulation (EC) No. 1881/2006), are generally higher in vegetables grown in heavily polluted soils (with a lead content above the critical immission value) compared to those grown in soil with lead contents below the critical immission values (Kirinčič et al. 2017). Furthermore, the authors found out that the average Pb concentrations are higher in the root vegetables than in the leafy vegetables, which are both most problematic if produced in critically polluted soil. Residents of the Upper Meža Valley, who consume leafy and root vegetables from their own gardens or fields, are therefore at increased risk of lead exposure, with children being particularly vulnerable to high levels (Kirinčič et al. 2017). This is why the local NIJZ unit actively promotes awareness of safe gardening and home-grown food consumption practices. They regularly share education materials, such as lectures, posters, leaflets and videos (NIJZ 2025) that emphasize basic safety measures. These measures include avoiding the growing of broad-leaf vegetables (e.g. lettuce and spinach) and tubers (such as beetroot, carrots and potatoes), replacing contaminated garden soil and thoroughly washing home-grown produce under sufficient running water before consumption. In the Žerjav area, which is the most polluted zone, a safe garden (with raised beds inside a greenhouse) was established. This garden has an important social

Table 2 Basic statistical data of Pb contents (mg/kg) in soil from the Upper Meža Valley, compared with previous studies

Parameter	Unit	This study									Pokorny et al. (2002)			Jež and Leštan (2015)		
		Garden & lawn soil			Garden soil (0–20 cm)			Lawn soil (0–5 cm)			Garden soil (0–20)			Garden, grassland and orchard soil (0–8, 8–14, 14–21 cm)		
		Č	M	Ž	Č	M	Ž	Č	M	Ž	Č	M	Ž	Č	M	Ž
Count		46	30	20	28	21	12	18	9	8	9	9	2	33	30	16
Minimum	mg/kg	23	54	75	140	54	75	23	170	310	511	573	3350	181	116	1331
Maximum	mg/kg	1600	2100	5100	1600	2100	4483	1200	1510	5100	1950	2830	4470	4517	3270	7384
Arithmetic mean	mg/kg	584	673	1397	659	667	1260	468	686	1604	1180	1090	3910	1267	1037	3892
Median	mg/kg	555	543	645	590	495	730	480	620	635	925	936	3910	1050	671	3789
Geometric mean	mg/kg	473	491	782	545	482	681	380	512	963	/	/	/	/	/	/

Č = Črna, M = Mežica, Ž = Žerjav; In bold are values used for modelling

and community function, where locals demonstrate the safe cultivation of vegetables and herbs. Compared to the median background value of 34 mg/kg for Slovenian soils (Gosar et al. 2019), the median Pb content in garden & lawn soil is enriched by 16 times in Črna and Mežica and by 19 times in Žerjav (Table 2). The geometric mean of Pb contents of all (lawn + garden) soil (Table 2) was used for modelling, since soil Pb contents typically follow a log-normal distribution with outliers (high Pb contents at some locations). The presence of highly skewed data distorts the arithmetic mean, making it less representative of the typical contamination levels in the area. The geometric mean therefore provides a more representative central value, reduces the influence of extreme outliers and aligns with the best practices in environmental geochemistry (Reimann et al. 2008). This is particularly relevant in our study, where a misleadingly high arithmetic mean, value could lead to misinterpretations of contamination severity.

The basic statistical data on Pb concentrations in drinking (tap) water and Pb contents in household dust are presented in Table 3. In Črna, all the tap water measurements ($n=4$) were above the LOQ value, while at monitoring locations in Mežica and Žerjav they were below the LOQ value. For this reason, the middle bound level of 0.5 µg/L was used for modelling in the case of Mežica and Žerjav. In the case of Črna, arithmetic mean was applied. All measured Pb concentrations in drinking water were below the parametric value for Pb of 5 µg/L set by Directive (EU) 2020/2184.

The median Pb contents in the house dust, determined in our study (Table 3), are approximately 4 times lower in Črna and 3 times lower in Mežica, than in the previous study focussing on the Upper Meža Valley (Flis et al. 2002; Table 3). Flis et al. (2002) collected samples in 2002, prior the Program of Measures. The Pb contents in house dust from Žerjav, as determined in this study, remain high. This is primarily due to the fact that Žerjav is the most heavily

Table 3 Basic statistical data of Pb contents (mg/kg) in household dust and of Pb concentrations (µg/L) in tap water, compared with previous studies

Parameter	Drinking water (µg/L)			Household dust (mg/kg)					
	This study			This study			Flis et al. (2002)		
Study area	Črna	Mežica	Žerjav	Črna	Mežica	Žerjav	Črna	Mežica	Žerjav
count	4	4	4	6	8	4	5	8	2
minimum	1.2	< 1	< 1	176	31	75	724	524	2126
maximum	2.7	< 1	< 1	344	829	2636	1426	1287	2277
arithmetic mean	1.775			266	319	1855	1089	896	2202
median	1.6			275	211	2354	1092	813	2202

In bold are values used in modelling

Table 4 Arithmetic mean of outdoor PM₁₀ Pb concentrations (ng/m³) in Črna, Mežica, Žerjav and Slovenia (rural and urban areas)

Study area	Location name	GKY ¹	GKX ¹	factor	Arithmetic mean Pb (µg/m ³)	Study area arithmetic mean Pb (µg/m ³)
Žerjav	<i>Permanent monitoring site</i>	490,348	149,042	1	0.3576	
	Žerjav 77	490,506	149,048	0.25	0.0894	0.0918
	Žerjav 20	490,465	149,131	0.32	0.1144	
	Žerjav market	490,584	149,025	0.2	0.0715	
Črna	Črna	488,923	147,890	0.2	0.0715	0.0805
	Črna	489,239	148,041	0.25	0.0894	
Mežica	Mežica	489,067	152,850	0.2	0.0715	0.0715
Slovenia urban area	Celje hospital	520,614	121,189	–	0.007	0.0084
	Ljubljana Bežigrad	462,673	102,490	–	0.0081	
	Ljubljana Biotehniška	459,457	100,591	–	0.008	
	Maribor Titova	550,305	157,414	–	0.00104	
Slovenia rural area	Iskrba	489,292	46,323	–	0.0027	0.0027

¹Spatial coordinate according to D48—Slovenia (ESRI:104,131)

²Data source for Upper Meža Valley (Slovenian Environmental Agency Air Quality archive) and for Slovenia (Koleša 2023)

In bold are values used for modelling

contaminated area, a legacy of historical smelting operations. Compared to the national baseline levels for household dust determined for rural (54 mg/kg) and urban (61 mg/kg) areas (Teran 2020), the median Pb contents in household dust from Črna, Mežica and Žerjav (Table 3) are 5 times, 4 times and 38 times higher, respectively.

The average yearly PM10 Pb contents in outdoor air for the study areas (Table 4) are below the regulatory limit of 0.5 µg/m³ (Official Gazette No. 9/11, 8/15, 66/18 in 44/22—ZVO-2). However, when compared to the PM10 Pb values determined for the Slovenian urban and rural areas (Table 4), the Pb contents in PM10 particles from the Upper Meža Valley are up to 11 times and 30 times higher, respectively.

Dietary Exposure and Exposure from Food Contact Ceramics

The estimated overall LB, MB and UB chronic dietary Pb exposure values for different scenarios are presented in Table 5. From Table S1 it is seen that relatively high shares of local food samples exceeded maximum levels of the Commission Regulation (EU) 2023/915 (EC, 2023), from 13.8% for Leaf vegetables to 92% for Milk and dairy products, which is not the case for the foods from the Slovenian market. In this study, the LB, MB and UB dietary exposure estimates for Pb in an average Slovenian toddler (1–3 years old) consuming only market-sourced foods were 0.75, 1.37 and 2.11 µg/kg bw/day, respectively (Table 5). These values can be compared to those from a previous study (Kirinčič et al. 2019), which reported 0.48, 0.95 and 1.42 µg/kg bw/day and an EU study (EFSA 2012), with values of 1.10, 1.32 and 1.54 µg/kg bw/day. The higher estimated exposure values in this study compared to the previous study (Kirinčič et al. 2019) are due to the inclusion of more food categories and the use of Slovene specific consumption data, which was not the case in the previous study. The estimated exposure values from this study fall within the range of values reported

in the EU study (EFSA 2012). Compared to the IEUBK default values of 5.21 µg/day (24–36 months), and 5.38 µg/day (36–48 months), which, when accounting for the average Slovenian toddler body weight of 12.7 kg (SI-MENU 2018), corresponds to 0.41 and 0.42 µg/kg bw/day, respectively, the LB arithmetic mean dietary exposure values for toddlers in this study (Table 5) were nearly twice as high in scenario D1, approximately five times higher in D2 and about eight times higher in D3. The dietary exposure estimates for general toddler population in this study (D1) are comparable to those from the EU but significantly higher than the IEUBK default values. Therefore, we replaced the latter with the dietary exposure estimates from this study. Since most food samples from the Slovenian market (Table S1) were non-detects, which could lead to a significant overestimation of the estimated MB and UB exposure, we decided to use the LB estimated arithmetic mean dietary exposure values in the modelling. The dietary exposures levels of toddlers (Table 5) all exceed the benchmark dose level for developmental neurotoxicity (BMDL01) set for Pb in children up to 7 years of age (0.50 µg/kg bw/day) (EFSA 2010b), indicating a potential health concern. Since no threshold has been established for several critical health effects of Pb, no tolerable dietary intake level has been recommended (EFSA 2010b).

Regarding the food contact ceramics, the distribution of samples and three different mean concentrations of lead in the simulant are presented in the supplementary material (Table S3). The mean concentrations ranged from 0.0057 to 0.016 mg/kg simulant, depending on how the data below the detection limit were censored. For all Slovenian samples, the migration of lead was below the migration limits pursuant to Directive 84/500/EEC (EC 1984). The exposure levels for the different scenarios are presented in Table 6. Exposure to lead from food contact ceramics was 0.018 µg/kg bw/day for the worst-case scenario (UB scenario). Exposure to lead migrating from the ceramic food contact materials represents less than 1% of exposure related only to dietary exposure

Table 5 Dietary lead exposure values according to the different dietary scenarios

Toddlers (1–3 years) dietary exposure	LB	MB	UB
	µg/kg bw/day		
D1: children consuming only Slovenian market foods (general reference scenario)	0.7	1.4	2.0
D2: most consumed food is from the market, except for half of the intake from specified local foods listed in Table S1	2.0	2.6	3.2
D3: children consuming specified local foods instead of market foods listed in Table S1, with the remainder sourced from the market	3.3	3.8	4.4
Market food and only milk and milk products from local produce	2.3	2.9	3.5
Market food and only eggs from local produce	1.0	1.6	2.3
Market food and only vegetables from local produce	1.3	1.9	2.5
Market food and only brook trout from local rivers	0.8	1.4	2.0

Overall chronic dietary exposure was estimated with 3 different arithmetic mean concentrations of Pb in different food groups: *LB* lower bound, *MB* middle bound and *UB* upper bound; the exposure scenario (D1, D2 and D3) values used for IEUBK modelling are shown in bold font—transformation to a unit of µg/day was based on a body weight of 12.7 kg (SI-MENU 2018)

Table 6 Exposure to lead (in $\mu\text{g/kg bw/day}$) from food contact materials for different scenarios and their parameters used in the second step of deterministic approach, together with the comparison between exposure to lead from ceramic products and exposure through food for A—the Slovenian population according to dietary exposure

Scenario Number	Allocation for hollowware	Substitution	Decreasing factor	Exposure from food contact ceramics ($\mu\text{g/kg bw/day}$)	% of D1	% of D2	% of D3
FCM1	0.85	LB	0.2	0.0038	0.50	0.19	0.12
FCM2	0.85	MB	0.2	0.11	0.77	0.41	0.28
FCM3	0.85	UB	0.2	0.018	0.89	0.56	0.40

FCM food contact material; D1, D2 & D3 = dietary exposure scenarios as presented in Table 5

scenarios D1, D2 and D3, which were established in this study. Since exposure to lead, migrating from ceramics into food simulants, represents less than 1% of the exposure related solely to dietary sources, such exposure was considered less significant and was not included in the IEUBK model.

Blood

The boxplots of the measured BLLs in children (2011–2022) from the case study areas are presented in Fig. 2. (Group 1) and Fig. 3 (Group 2). For both groups,

most children living in these areas have had BLLs exceeding the blood lead reference value (BLRV) of $35 \mu\text{g/L}$, which was established by the Center for Disease Control and Prevention (CDC) in 2021 (US CDC 2024) as a threshold for identifying children with elevated BLLs compared to the general US population. Compared to the BLL of $50 \mu\text{g/L}$, the level at which there is sufficient evidence of adverse health effects in children and adults (WHO 2021; ATSDR 2024), concerning levels of BLLs were found in approximately three quarters of children from Žerjav, and about half of the children from Črna and Mežica. The interquartile ranges (IQR ranges = Q1—Q3)

Fig. 2 Comparison of the observed blood lead levels (BLLs) (boxplot distributions) with the estimated BLLs (point values) according to the established dietary scenarios (D1, D2, D3 and IEUBK default value) for group 1 (24–36 months); the y-axis was cut off at $350 \mu\text{g/L}$ for better visualization

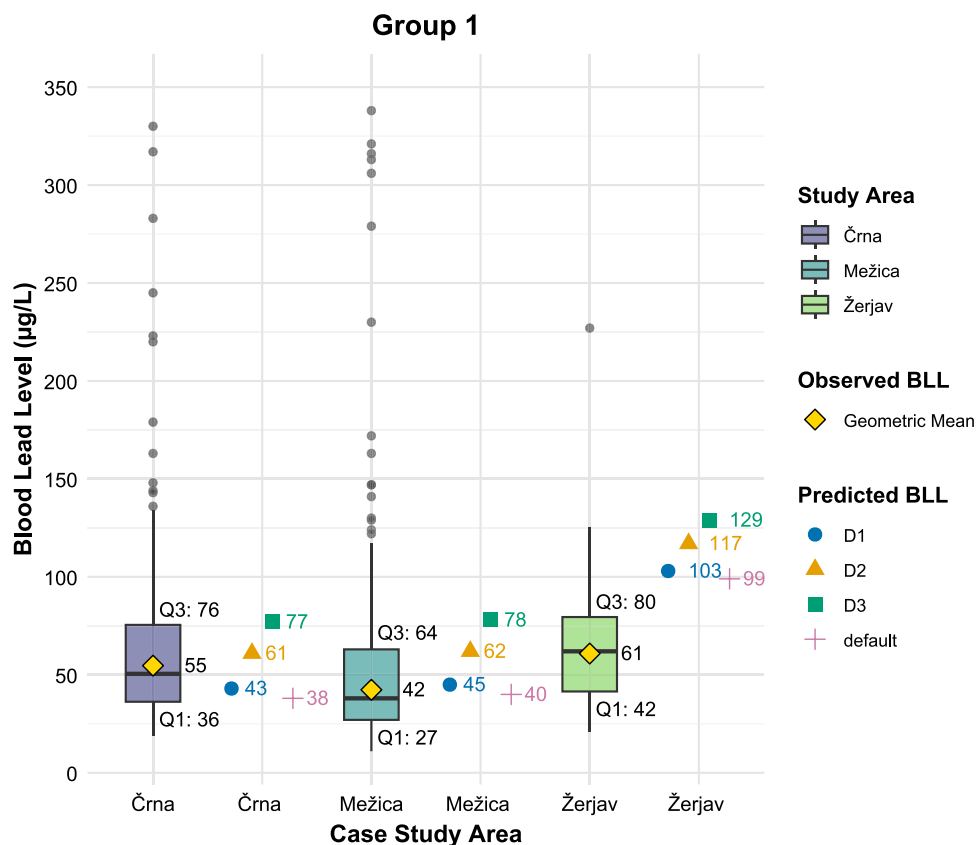
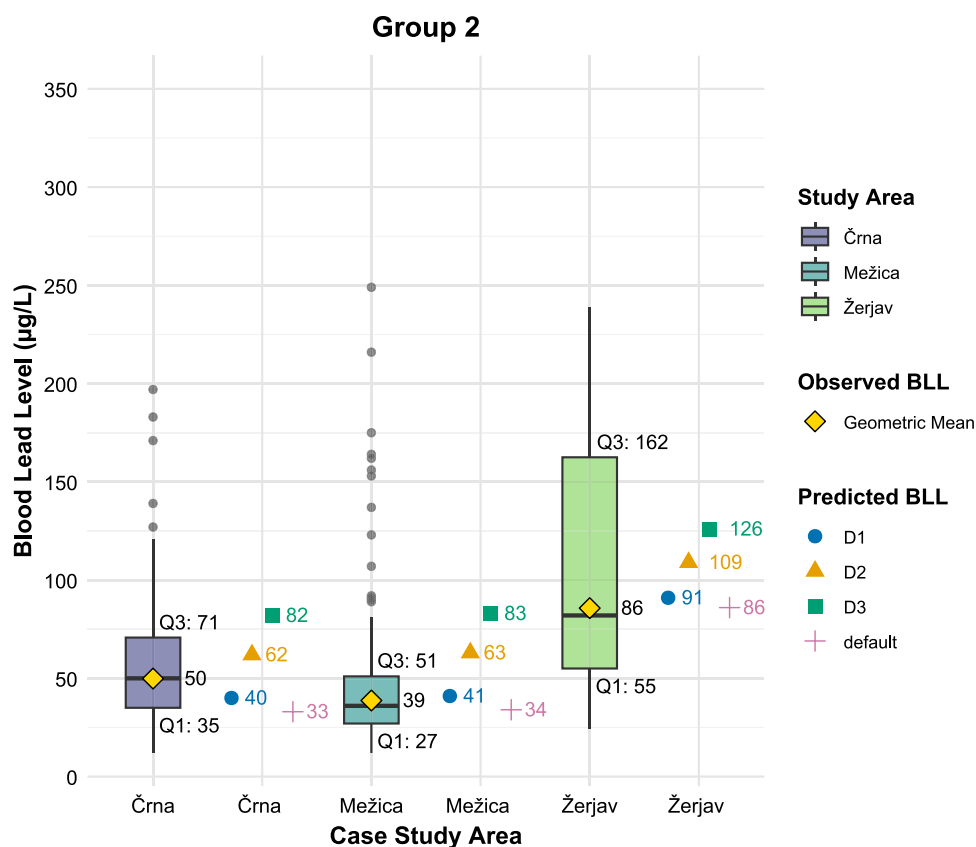


Fig. 3 Comparison of the observed blood lead levels (BLLs) (boxplot distributions) with the estimated BLLs (point values) according to the established dietary scenarios (D1, D2, D3 and IEUBK default value) for group 2 (36–48 months); the y-axis was cut off at 350 µg/L for better visualization



of the measured BLLs, which are shown in Figs. 2 and 3, were defined as general exposure profiles for the studied areas.

Pb exposure Modelling

Upper Meža Valley

The results of modelling are provided in Table S4. If the estimated BLLs for different dietary exposure scenarios fell within the interquartile range (IQR) of the observed BLLs (Figs. 2 and 3), they were considered to be in reasonably good agreement. This was the case for the Črna area under scenarios D1 and D2 for both groups, and for the Žerjav area under all scenarios for group 2. In Mežica, scenarios D1 and D2 aligned well with the measured BLLs in Group 1. However, for Group 2, scenario D2 slightly exceeded Q3. Models based on scenario D3 tended to overestimate the exposure in both groups in Mežica and Črna. This suggests that these models better match children with high (above Q3) or extreme (outliers) measured BLLs. A similar pattern was observed for all models in Žerjav for Group 1. Considering the geometric means of observed BLLs for both groups (Figs. 2 and 3), the findings were as follows: In the Mežica area, the geometric mean closely aligns with the predicted BLL based on the scenario D1 (assuming that only market

foods are consumed). In Črna, it falls between the predicted BLLs for scenarios D1 and D2 (assuming consumption of only market foods, and to some extent local foods). In Žerjav (Group 2), it is identical to the predicted BLL based on the IEUBK default diet value. For Group 1, however, the observed geometric mean was significantly lower than all the predicted BLLs.

Regarding the relatively high predicted BLLs for Group 1 in the Žerjav area, it should be noted that fewer BLLs (Group 1: 14; Group 2: 18) were observed for both groups in Žerjav compared to Črna (Group 1: 158; Group 2: 142) and Mežica (Group 1: 250; Group 2: 204). This discrepancy is largely due to the very low number of children living in Žerjav area. Between 2008 and 2023, the number of children aged 0–4 years living in Žerjav fluctuated between 7 and 13 per year. This extremely small and fluctuating population base introduces several analytical challenges, such as increased sampling error, reduced capacity to detect significant differences and potential for unrepresentative sampling. These factors collectively limit the reliability of any cross-area comparisons and suggest caution in interpreting the Žerjav results as indicative of true exposure patterns. With only 14 children in the Group 1 validation dataset, statistical variability is high, so a small sample size may not capture the true observed BLL variability and may bias the results towards lower observed values. However, the relatively high

BLLs predictions for both groups align with scenario D3 for Črna and Mežica. These relatively high BLLs are likely due to several factors, including possible errors or uncertainties in the measured environmental lead concentrations and exposure pathways (e.g. dietary lead), which could contribute to overestimation. The combination of small sample sizes and measurement uncertainties creates a compounded error structure that should be acknowledged when interpreting these predictive models. The uncertainty in the dietary exposure estimates arises from using the same mean lead levels for local foods across all 3 studied areas due to the insufficient sample data. It has been observed that certain local food products in the Žerjav area exhibit the highest Pb concentrations (Ribarič-Lasnik et al. 2002). In Žerjav, Pb levels in household dust were based on limited (4 samples) and outdated Pb dust measurements (from 2011 and 2016) showing high values (Table 3). Additionally, the household dust was not collected from the homes of children, who participated in the blood analyses, as was done in Črna and Mežica, where such sampling was conducted.

As for Group 1 in the Žerjav area, physiological differences in children aged 24–36 months compared to the model's default parameters (derived from U.S. children) could contribute to further discrepancies. If the uptake or biokinetic assumptions do not accurately reflect local physiology, the predicted BLLs may be overestimated. In addition, default IEUBK assumptions about behavioural factors such as soil and dust ingestion rates may not be representative of children in Žerjav. For example, outdoor activity patterns and hand-to-mouth behaviour may differ due to local cultural or environmental conditions, potentially resulting in lower actual ingestion than modelled. Similarly, higher assumed ingestion rates and bioavailability of lead from dust and soil could result in inflated prediction of lead absorption. For soil, the assumed bioavailability appears reasonable, as site-specific values (Jež & Leštan 2015) across locations

(~25–28%) were comparable to the IEUBK default value of 30%. Regarding house dust, the same ingestion rate and bioavailability were applied in all three areas (Črna, Mežica, Žerjav) and the estimated and measured BLLs generally matched quite well. However, the uniform application of dust bioavailability and ingestion rates across all three areas may not reflect the true exposure in Žerjav. In particular, if actual dust exposure is lower due to housing conditions or behavioural factors, the model may overestimate internal dose.

The results of the contribution of the different sources to the overall Pb exposure for Groups 1 and 2, which are based on the different dietary exposure scenarios (Figs. 4 and 5), are similar. Therefore, only the results for children in age Group 1 (24–36 months) (Fig. 4) are discussed further. The analysis shows that soil/dust is a major exposure pathway in Žerjav, while diet contributes most of the remaining exposure. In Črna and Mežica, soil/dust is the dominant exposure pathway only in the case of scenarios D1. For scenarios D2 and D3, which include local food consumption, dietary exposure becomes the primary pathway, with soil/dust contributing the remainder. Drinking water and air account for less than 1% of total exposure in all areas, except in Črna, where drinking water represents up to 5%. If the IEUBK default diet values were used in the modelling, the dietary contribution would decrease to 25.2% in Črna, 24.2% in Mežica and 8.6% in Žerjav. This demonstrates that relying on the IEUBK default diet values may underestimate the role of dietary exposure.

In this study, dietary exposure in the Upper Meža Valley was studied in detail for the first time. In the first application of the IEUBK model in the Meža Valley (Ivartnik and Eržen 2010), using the default diet values, it was found that in all three areas (Mežica, Črna and Žerjav), 86.6–96.8% of lead uptake came from soil and dust, 3.4–4.8% from drinking water and 0.06–0.15% from air. Since local dietary exposure

Fig. 4 The contribution of sources to overall lead exposure (in %) for group 1 (24–36 months)

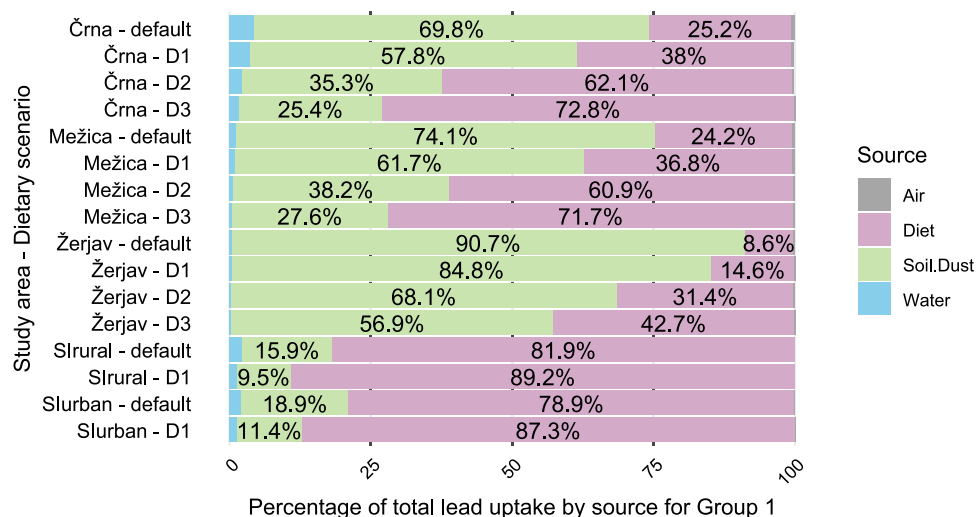
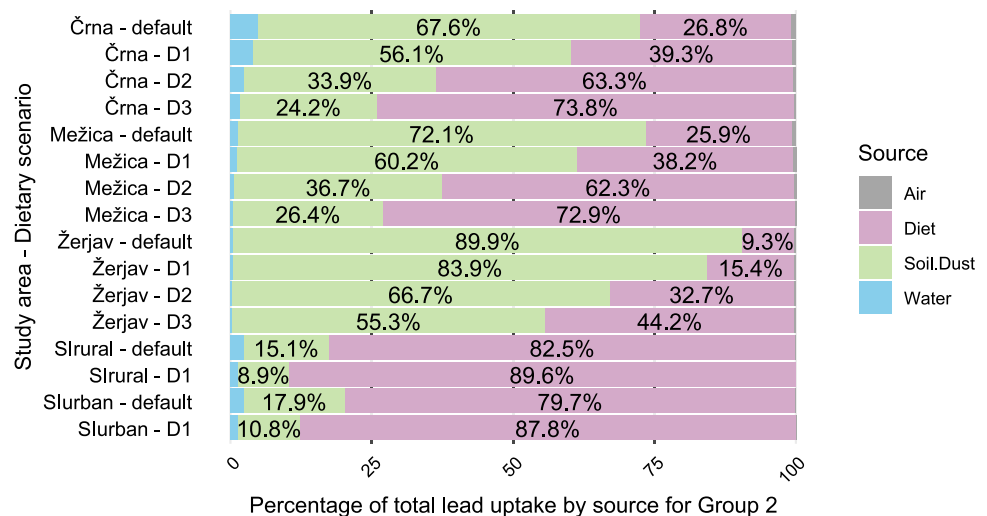


Fig. 5 The contribution of sources to overall lead exposure (in %) for group 2 (36–48 months)



data were unavailable, the contribution of diet to lead intake was not assessed. In our study, only in the Žerjav area (based on a scenario using either the default diet value or market food value (D1)) was the Pb share close to the previously determined range (Ivartnik and Eržen 2010). Furthermore, in the Mežica and Črna areas, dietary exposure is the major exposure pathway in the scenarios, where the diet includes consumption of local foods. This can be explained by the following reasons. First, the site-specific dietary exposure values in our study are notably higher than the IEUBK default value. Additionally, the site-specific soil and dust levels in the study by Ivartnik and Eržen (2010) were much higher, as the measurements were taken in 2002 (smelting operations closed in 1994), before the Program of Measures was implemented in the Upper Meža Valley. It is also assumed that the comprehensive measures implemented between 2007 and 2022 have significantly reduced the environmental Pb exposure. This has already been shown in the bio-monitoring program (Ivartnik et al. 2021). In addition, Jež and Leštan (2015) demonstrated that the soil remediation (washing with EDTA) could reduce the Pb contents of soil in the areas of Mežica, Žerjav and Črna by 53%, 67% and 62%, respectively, while also decreasing the concentration of in vitro bioaccessible Pb in the simulated human gastric phase by 2.6, 3.2 and 2.9 times, respectively. The results of IEUBK modelling indicated that after soil remediation, the number of locations where the expected BLL in children exceeded the stipulated 100 µg/l would decrease by 90%, 38% and 91%, respectively. Consequently, soil remediation was advised for Črna and Mežica, whereas in Žerjav, soil capping or removal was advised due to the limited improvement in predicted BLLs after remediation. This is because the Žerjav area was built around the central former Pb smelting plant, where soil pollution is the most severe (Finžgar et al. 2014). Jež and Leštan (2015) also emphasized that consuming of home-grown vegetables could be a significant

source of Pb intake, as most households in the Meža Valley cultivate vegetable gardens. Although their study did not determine site-specific dietary values (relying instead on default values), our findings support their assumptions.

In studies applying IEUBK modelling at brownfield sites impacted by mining, particularly smelting activities, the default IEUBK diet values are rarely replaced by the site-specific values. Carrizales et al. (2006) used a site-specific dietary exposure value of 9.3 µg/day (equivalent to 0.55 µg/kg bw/day, assuming a mean body weight of 17 kg, calculated from median standard values from EFSA (2013)) in a city of San Luis Potosi, (Mexico), where a copper-arsenic (lead) smelter operates. Their findings indicated that soil and dust accounted for 87% of the BLLs in children aged 3–6-year. Zhang et al. (2017) investigated a historic mining and smelting town in south China and determined that, for 3–4-year-old children (GM BLL = 87.4 µg/L) 87.6% of Pb comes from soil and dust, 8% from diet, 4% from water and 0.4% from air. Compared to the dietary exposure data in our study, they used site-specific average lead contents for a limited number of home-grown foods (rice, vegetables, chicken and pork), which represents a much smaller list of consumed foods. Although site-specific eating habits were considered in their study, it was found that the consumption of home-grown foods only accounted for 17% of the total diet due to the limited farmland in the studied region. Li et al. (2016) found out that in children aged 61–84 months living near a battery factory and near a lead–zinc mine, on average 83.39% (57.40–93%) of BLL were attributed to diet, while 15.18% (3.25–41.60%) came from soil/dust exposure, with the remainder contributed by air and drinking water. The site-specific dietary Pb intake values in their study were even higher than in ours (ranging from 35.33 to 77.63 µg/day, equivalent to 1.77–3.88 µg/kg bw/day, assuming a mean body weight of 20 kg, calculated from median standard values from EFSA 2013). These values were based

on measurements of staple foods and vegetables collected from the school canteen meals, where the recruited children ate three meals per day. Since the food samples were not collected from the local farmlands, the local food contribution was not investigated. However, the authors suggest that the high Pb contents in staple foods and vegetables may be partly due to the Pb contamination of local farmland soil. Besides the site-specific data for all exposure media, they also incorporated specific data on outdoor and indoor activity time, ventilation rates and water consumption, as evaluated through questionnaires. Both our study and the study by Li et al. (2016) show that the dietary exposure source in mining or industrial contaminated areas can be quite significant, particularly due to the consumption of locally produced foods. This highlights that relying on the default diet values in the IEUBK model for such areas may underestimate the dietary Pb exposure contribution.

Tragically, the Upper Meža Valley was severely affected by a catastrophic flooding in 2023 (Mikoš and Sodnik 2024), which left behind destroyed roads, bridges and residential buildings while depositing hazardous materials (debris, sediment, etc.) onto inhabited areas. The floods inundated buildings, infrastructure and flood plains, including some agricultural land. Considered the worst natural disaster in the country's history, the event drastically diminished the effectiveness of past mitigation measures in the Upper Meža Valley. In our study, the determined site-specific values are based on pre-flood measurements. As such, this study provides a comprehensive assessment of baseline environmental lead contamination and lead exposure levels in children's (24–48 months old) before the flood's impacts. Consequently, it serves as a valuable reference for evaluating potential flood-related effects on Pb contamination and human exposure. The devastating 2023 flood in the Upper Meža Valley necessitates future research focussed on assessing the post-flood environmental lead contamination in soil, dust, sediment, water and local foods, and comparing it to the pre-flood baseline established in this study. Crucially, follow-up biomonitoring of children's blood lead levels and re-evaluation of exposure pathways are needed to determine the impact of the flood on human exposure. Furthermore, research should investigate the effectiveness of past mitigation efforts, model the redistribution of lead and conduct long-term health outcome studies to inform future risk assessments and mitigation strategies in the affected communities.

Comparison with the Slovenian General Children Population The IEUBK model was used to assess Pb exposure in the general children's population. For Group 1, the estimated BLLs for an average Slovenian child were 19 µg/L in rural areas (based on scenario LBD1) and 20 µg/L in urban areas. In Group 2, the estimated BLLs were 20 µg/L for both rural

and urban areas. No national biomonitoring data are available for validation in the discussed age groups. However, a geometric mean BLL of 13.4 µg/L (range of 6.9–24 µg/L) was reported for children aged 7–11 years in Slovenian urban areas (Hrubá et al. 2012). When comparing the modelled values with measured BLLs in children from other EU developed countries, the estimates appear realistic. Examples include an arithmetic mean BLL of 21 ± 0.6 µg/L in 1–3-year-old children and of 15.9 ± 0.6 µg/L in 4–11-year-old children from Almería (Spain) (Ruiz-Tudela et al. 2021) and a geometric mean BLLs of 15.2 µg/L for 2–3-year old children and of 15.0 for 3–4-year-old children from France (Etchevers et al. 2014; 2015).

The results show that most children from the Upper Meža Valley had higher values than the average Slovenian child from rural and urban areas. On average, in Group 1, the observed BLLs were about twice as high in Mežica and Črna and three times as high in Žerjav, when compared with the estimated BLLs of average Slovenian child's BLL value of 19/20 µg/L, with the medians of the observed values in Črna (51 µg/L), Mežica (38 µg/L) and Žerjav (62 µg/L). In Group 2, the observed BLLs were approximately twice as high in Mežica and Črna and about four times higher in Žerjav, compared to the estimated average Slovenian child's value of 20 µg/L, with the medians of the observed values in Mežica (50 µg/L), Črna (37 µg/L) and Žerjav (82 µg/L).

The results regarding the contribution of the different sources to the overall Pb exposure in the general population of Slovenian children (Figs. 4 and 5) indicate a negligible difference (between 1 and 2%) between age groups 1 and 2 for each source. In both rural and urban areas, diet was identified as the major exposure pathway (89.2 and 87.3%, respectively), followed by soil/dust (9.5 and 11.4%, respectively). Drinking water accounted for approximately 2% of exposure, while air contributed less than 1% in both areas. If the IEUBK default diet values were used instead of D1 (derived from the Slovenian market), the dietary exposure contribution would decrease, with a corresponding increase of approximately 7% in the soil/dust share.

Conclusion

This study utilized a comprehensive, site-specific and most up-to-date dataset encompassing environmental, dietary and biomonitoring dataset, along with expert knowledge, to improve exposure assessment from multiple sources and routes in a historic mining area. Lead exposure in children (24–48 months) was thoroughly investigated. Site-specific soil and locally sourced food Pb concentrations significantly exceeded regulatory limits, raising substantial health concerns. Compared to the national baseline levels, the Pb contents in household dust are also high and thus require

particular attention. Tap water concentrations and outdoor air contents were found to be within acceptable limits. If the site-specific environmental source values determined in this study represent a reliable general situation in the studied areas, the findings suggest that even in a relatively highly contaminated Pb mining area, dietary exposure can be a significant source of Pb in children. The results of the study indicate that the comprehensive measures, implemented under the Program of Measures, successfully reduced the environmental exposure, particularly in the areas of Mežica and Črna areas. However, in Žerjav, environmental burden is so severe that even these measures were not fully effective. Since a diet high in locally sourced foods may pose a significant exposure risk, ongoing attention must be on the dietary habits of children living in the Upper Meža Valley. Since the implementation of Program of Measures in the Upper Meža Valley, steps have been taken to exclude locally produced foods from children's meals in kindergartens. Residents have also been advised to limit their consumption of lead-accumulating foods, particularly root and leafy vegetables, grown in the area. Preventive measures concerning environmental and dietary exposure were discussed in detail regularly with those families, whose children had BLLs exceeding 100 µg/L. In conclusion, the findings of this study provide further evidence that the Upper Meža Valley remains a long-term Pb contaminated residential area, requiring ongoing efforts to minimize children exposure, particularly to soil, household dust and local dietary sources of Pb. These efforts are now a high priority, as the catastrophic floods in 2023 nullified some of the previously implemented measures. Exposure through drinking water, air and food contact ceramics is not of particular concern.

Uncertainties and Further Research

The lead values in the 2023 house dust data used in this study were obtained through voluntary participation of residents in the Upper Meža Valley. However, the reported Pb contents in household dust are likely underestimated, as participants in this study were more environmentally conscious and proactive about local pollution issues. To account for this potential bias, we incorporated some data from previous studies, where Pb contents were significantly higher, providing a more realistic assessment. We also recognize that dust collection methods, such as using participants' vacuum cleaners, introduced variability due to differences in vacuum cleaner models, cleaning habits and collection times, which may have contributed to heterogeneity in the Pb concentrations (Gillings et al. 2022). To refine the IEUBK model, future research will focus on dust bioaccessibility and comparison between house dust and nearby soils. Alongside dust sampling, we will distribute questionnaires to participating

households to gather data on human activity patterns. This information will be integrated into further analyses when comparing measured and estimated exposure values for individuals.

The number of samples measuring lead concentrations in drinking water was relatively low. However, uncontaminated karst aquifer groundwater is the main source of tap water in the Upper Meža Valley. Therefore, it is assumed that the drinking water in Upper Meža Valley is generally of good quality, as indicated by the public monitoring programs.

Regarding the uncertainties in the dietary exposure assessment, the Pb food content data were partly obtained from the targeted sampling of foods generally known to be the main source of Pb. This could lead to an overestimation of exposure. For food groups not included in the Slovenian database, EU mean contents were used, which could result in either overestimation or underestimation of exposure. Additionally, use of means calculated from datasets with a high proportion of non-detects may further contribute to underestimation or overestimation of exposure. Some uncertainty also arises from using the same mean lead contents of local foods for all 3 studied areas. Additionally, part of the uncertainty can be attributed to the differences in age groups considered in the dietary exposure assessment and the IEUBK model. In the dietary exposure assessment, toddlers 1–3 years old were treated as a single age group due to the available age classifications in the food consumption data (EFSA, 2024). However, the IEUBK model requires children between 1 and 3 years old to be modelled separately (24–36 and 36–48 months). The deterministic approach to dietary exposure assessment used in this study has an inherent tendency to produce conservative estimates (Ferrari et al. 2013), potentially leading to overestimation. Another limitation of this study is the lack of data on consumer habits related to home-grown or locally sourced vegetables and other foods, including the types and quantities consumed compared to market-bought foods. These habits vary depending on preferences and local conditions, significantly affecting dietary exposure estimates.

The annual number of children with elevated BLLs in all case study areas is relatively low, particularly in Žerjav. Notable differences in BLLs between the case study areas were found, especially in Žerjav, where BLLs are much higher compared to Mežica and Črna. While environmental factors and local based food dietary exposure are major contributors to higher BLLs (in Žerjav, this is largely due to a more severe environmental burden from historical smelting facilities in the area), also socio-economic status also plays an important role. Through questionnaires along with blood sample collections and home visits with families, whose children had BLLs above 100 µg/L, it was found that families with higher socio-economic status generally make greater efforts to reduce the Pb exposure. This highlights the

need for further study on the influence of socio-economic status on BLLs.

Exposure to lead from migration out of ceramics is likely overestimated, primarily due to the main uncertainties—the migration test method and the food consumption data used. Relying on regulatory lead migration data obtained through a food simulant, rather than actual data on migration measurements into food, introduces a degree of uncertainty. Additionally, the food consumption data used in this study are based solely on a worst-case scenario involving contact with fruit and vegetable juices and nectars, although other types of food may also come into contact with ceramics.

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Author Contributions All authors contributed to the study conception and design. Špela Bavec: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Resources, Writing—original draft, Writing—review and editing, Supervision and Visualization. Teja Čeruš: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Resources, Writing—original draft, Writing—review and editing, Validation and Visualization. Stanislava Kirinčič: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Resources, Writing—review and editing, Validation and Visualization. Matej Ivartnik: Conceptualization, Data curation, Investigation, Methodology, Resources, Writing—review and editing and Validation. Viviana Golja: Data curation, Formal analysis, Investigation, Resources and Writing—review and editing. Janja Turšič: Data curation, Formal analysis, Investigation, Methodology, Resources, Writing—review and editing and Validation. Klemen Teran: Data curation, Formal analysis, Investigation, Resources and Writing—review and editing. Miloš Miler: Data curation, Formal analysis, Investigation, Resources and Writing—review and editing. All authors have read and approved the final manuscript.

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Data Availability Due to the sensitive nature of the raw data (particularly the blood, soil and house dust data) and related geographic location (the identity of a child might be exposed in case of sharing

the coordinates), participants providing blood, soil and dust data were assured that the raw data would remain confidential. However, in case of interest to use these data (exclusively for scientific or expert purposes), authors of this study or the data managing institutions (Geological survey of Slovenia: house dust data; National Institute for Public Health: drinking water, diet, migration from food contact ceramics and blood data; Slovenian Environmental Agency: soil) should be contacted to help with the possibility to obtain raw data. Air PM10 data are publicly available (in Slovenian only) in the Air quality archive of Slovenian Environment Agency (<https://www.arso.gov.si/zrak/kakovost%20zraka/podatki/arhiv.html>).

Declarations

Competing Interests The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Ethical Approval The surveillance for human biomonitoring was approved by the National Medical Ethics Committee of Slovenia and a certificate of approval is available upon request. Each parent whose child participated in the biomonitoring program signed a statement agreeing to the blood withdrawal. All statements are kept in a locked cabinet at the National Institute for Public Health.

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