Health risk assessment of potentially toxic elements in the riparian zone of the Sava River (southeastern Europe): effects of high and low water events

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Abstract

This study evaluated the source apportionment and level of contamination with potential toxic elements (PTEs)—As, Cd, Cr, Cu, Ni, Pb and Zn—in a riparian zone of the Sava River by calculating the Enrichment Factor (EF) and the Geo-Accumulation Index (Igeo). The study analysed the non-carcinogenic and carcinogenic health risks to both adults and children in the local population. Soil and sediment samples collected during high and low water events from 12 sites along 945 km of the river were screened. Elevated soil contents of As, Cr, Cu, Ni, and Zn were observed along the middle and lower stretches from Županja (ZUP) to Belgrade (BEO), and an elevated Cu content was detected in the upper stretch Čatež (CAT), highlighting these elements as a potential risk for the human population. Elevated As, Cd Cr, Ni, and Zn levels in the sediment were also detected, with the Ni content three times higher than the Permissible Exposure Limit (PEL) values, indicating high contamination. The study established that As, Cr, and Ni primarily originate from geological sources; Cu, Pb, and Zn are predominantly from anthropogenic sources; and Cd originates from both geological and anthropogenic sources. Although the EF and Igeo varied along the entire river stretch, they were significantly higher for most elements in the lower stretch, indicating intensive anthropogenic contamination and potential health risks. The carcinogenic and non-carcinogenic risks associated with soils and sediments during high and low water events did not imply a potential risk for adults. However, As, Cd, Cr, and Pb potentially pose the greatest non-carcinogenic risk for children in both high and low water events, with ingestion as the main route of exposure. According to our findings, the elevated concentrations of As, Cd, Cr, and Pb in the Sava River basin (SRB) pose a significant potential health risk to the local population based on several factors: the high levels of these elements, their potential contamination risk as determined by the EF and Igeo, and their bioavailability and solubility. Although the cumulative Total Hazard Index (THI) and Total Carcinogenic Risk (TCR) values are low, particularly in the lower stretch of the Sava River, these elements still present a high potential health threat.

Keywords Sava River, Contamination risk, Human health risk, Potentially toxic elements, Enrichment Factor, Geo-Accumulation Index

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Introduction

Large rivers in Europe are affected by multiple stressors, such as water scarcity, urban, industrial, and agricultural land use, hydropower generation, and climate change [1]. This also applies to water-dependent riparian ecosystems. As river pollution can be transboundary, a

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continuous assessment of pollutants in water, sediments, biota, riparian soils, and vegetation is important. Riparian zones, defined as transitional areas between terrestrial and freshwater ecosystems, are characterized by distinctive soil, hydrological, and biotic conditions [2]. Due to the critical ecological role, fragility, and the valuable ecosystem services these environments provide to society, extensive research from various perspectives is essential.

Globally, floodplains offer a range of unique and essential ecosystem functions and services. These include water supply, flood risk reduction, maintenance of base flows, carbon sequestration, sediment retention, provision of food and recreational facilities, and serving as a source of biodiversity [3, 4]. Due to ever-growing demand, the sustainable provision of these services is increasingly under threat [5]. In the last century, human activities have caused riparian ecosystems to undergo more accelerated and extensive changes, primarily to meet the rapidly increasing demand for food, fresh water, timber, fibre, and fuel [4, 6].

The imbalance between the importance of these ecosystems for livelihoods and the environmental risks they face has resulted in large fluvial systems becoming some of the most degraded and threatened in the world. This has led to significant degradation, contamination, and a largely irreversible loss of biodiversity [7, 8].

Various anthropogenic activities such as urbanization, industrialization, deforestation, and the overuse of fossil fuels have contributed significantly to climate change, which has led to the intensification of natural disasters such as floods and extreme droughts. In recent years, flooding has been declared one of the most significant global challenges [9, 10]. Extreme flooding leads to widespread redistribution of pollutants, especially PTEs, across extensive areas, with long-term consequences for socio-economic, environmental, and agricultural sectors, which has been particularly noticeable in European countries in recent years [11, 12]. In general, PTEs are naturally present due to the weathering of parent material [9, 13]; however, anthropogenic activities such as mining and industrial processing of mineral resources and their subsequent use in industry and agriculture have significantly increased their concentrations in the environment [14]. Mining and industrial production release PTEs into rivers through atmospheric deposition and mining wastewater. Once in the aquatic environment, PTEs can quickly bioaccumulate, easily entering the human food chain and impacting human health [10, 15].

With an area of 97,700 km^2, the SRB is the largest catchment area in southeastern Europe. The Sava and its riparian zones are known for significant biodiversity and industrial and economic prospects. A large part of

the population in the SRB (about 3 million people, 16) lives close to the river banks and depends on it. In addition, the river is the primary source of drinking water for more than 9 million inhabitants [17]. The highest population density is near large cities, while agriculture dominates in the Croatian and Serbian parts of the catchment area. Thermal and hydroelectric power plants, oil and gas refineries, the metallurgical, chemical, and textile industries, and mining (heavy metals and salts) are the main anthropogenic influences in the catchment area [18]. The accumulation of potentially toxic elements (PTEs) in the soil and sediments of the Sava River's riparian zone, driven by human activities, poses a significant environmental and health threat because of their high toxicity, longevity, and tendency to bioaccumulate [19]. One serious problem in the SRB is extreme flooding caused by climate change. These events cause major socio-economic and health problems as PTEs can be transported over long distances during high water levels, primarily affecting agricultural land on which most of the population depends.

The most common routes of exposure to PTEs for both adults and children include ingestion, inhalation, and dermal contact [20–22]. Children are particularly vulnerable due to their weaker immune systems and specific hygiene behaviours. Although some elements like Zn, Cu, and Cr are considered vital for organisms, they could be toxic at high concentrations [23]; however, other elements such as Cd, Hg, and Pb have no useful functions for living organisms and are highly toxic even at low concentrations [24]. Due to excessive or long-term exposure, even low concentrations of As, Pb, Cr, or Cd can cause serious health problems such as cardiovascular diseases, skin lesions, reproductive and haematological damage, nervous system disorders, developmental anomalies, liver and kidney dysfunction, and skin and lung cancer [19, 25, 26].

The potential health risks that may result from over-exposure to PTEs should not be ignored. The main objectives of this study were to assess the level of contamination by calculating the Enrichment Factor (EF) and the Geo-Accumulation Index (Igeo) based on the content and origin of PTEs (As, Cd, Cr, Cu, Ni, Pb, and Zn) listed as priority substances in the EU Water Framework Directives, and to assess their impact on the health of the local population (children and adults) by analysing the non-carcinogenic and carcinogenic health risks. This study includes sites along the entire course of the Sava River. All parameters were analysed during high and low water events to assess whether the fluctuations in river levels affect human health. The data obtained in this study, based on widely recognized scientific methods, are applicable in defining potential risk zones where people,

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particularly children, are exposed to non-carcinogenic and carcinogenic risks from PTE pollution. These results can be strong arguments for developing pollution control and reduction plans, especially in settlements exposed to frequent flooding in the SRB and globally.

## Materials and methods

Study area

The Sava River, which is 945 km long, is one of the largest and most important tributaries of the Danube and its catchment area covers the territory of four countries: Slovenia, Croatia, Bosnia and Herzegovina and Serbia (Fig. 1). It partly encompasses the territory of Montenegro, with a minor portion of the basin extending into the territory of Albania [16, 27].

The SRB has a diverse topography and can be generally divided into three distinct regions: high mountains, low mountains, and lowlands. An important geological feature of the SRB is that it consists of very thick layers of limestone, complemented by sandstone, marl, mudstones, intrusive and extrusive igneous and metamorphic rocks in the rest of the basin [16]. Due to the diversity of the geological substrate, climatic characteristics, biodiversity, topographical and hydrological conditions, and even anthropogenic influences, different soil types have formed in the SRB. Soil formation in the riparian zone of the river is determined by erosion and sedimentation processes, changes in oxidation–reduction conditions,

frequent fluctuations in groundwater levels, and periodic floods [27]. In terms of soil composition, fluvisols dominate in the SRB, except in the uppermost section, where undeveloped skeletal soils predominate [28–30]. A considerable part of the basin is covered by forests and semi-natural areas (54.7%), and agricultural areas (42.4%) [16]. Human-made features, such as discontinuous urban zones, road and rail networks, mining areas, and landfills, cover less than 3% of the total land area of the basin [16].

The climate across the basin varies due to orographic features, with two main types identified: alpine climate and moderate continental climate. The average annual temperature for the whole SRB is 9.5 °C [16], and the average amount of precipitation for the basin is about 1100 mm. The spatial distribution of the runoff in the SRB follows the spatial distribution of precipitation. The annual runoff in the basin varies between 150 mm, which corresponds to less than 5 l/s/km^2, and almost 1200 mm per year, which corresponds to about 40 l/s/km^2. The highest rainfall occurs in early summer and autumn, when flooding is most frequent. Spring floods are much longer and less intense, while autumn floods are shorter but more extreme [3].

Riparian zones are of interest because of their high organic residue but low humus content, influenced by annual sediment deposition from river floods that disrupts decomposition processes [31]. Consequently, these zones have unique hydrological and soil characteristics
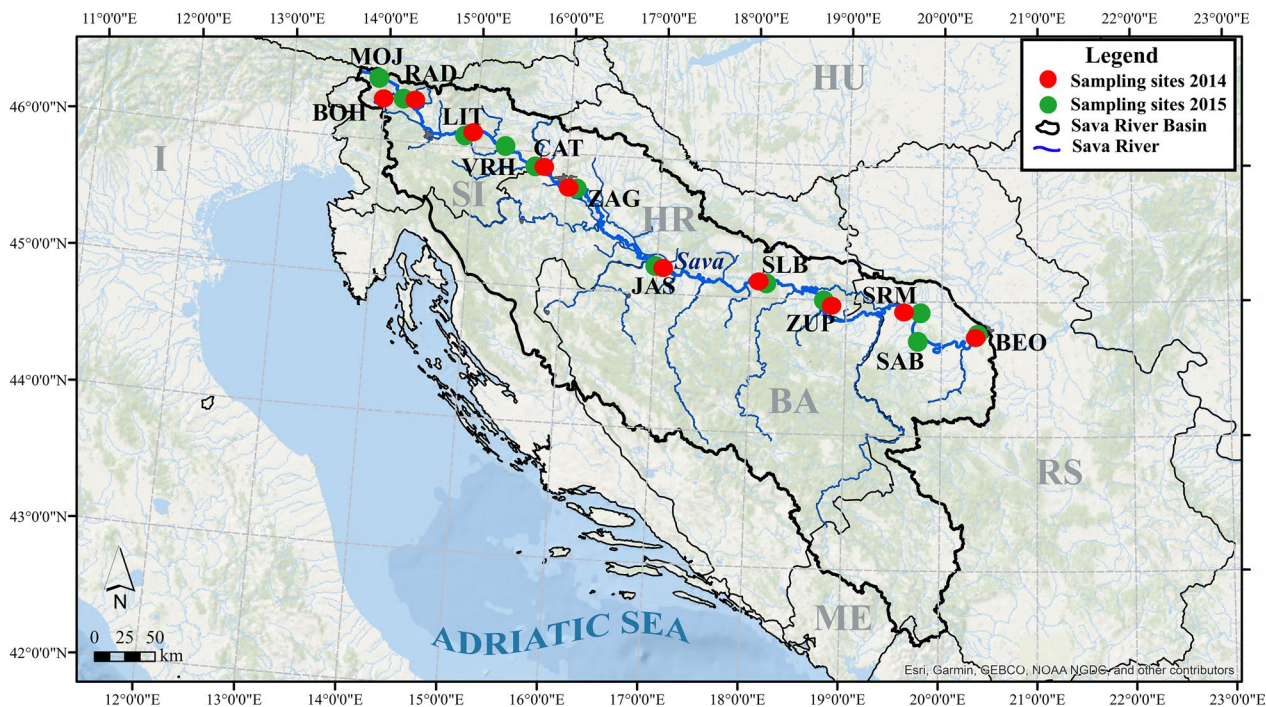
Fig. 1 Sampling sites along the Sava River

associated with a wide diversity of vegetation. Extensive industrialization and urbanization of the SRB impact the river, its ecological state, and its biodiversity. The sources of pollution in the riparian zones of the Sava River are mainly related to chemical weathering of parent material, the release of industrial waste, untreated wastewater from municipalities, and pollutants from agricultural activities [32]. The spatial gradient shows that the upper reaches are mainly influenced by the weathering of carbonate minerals and ore deposits, the middle reaches by agricultural activities (the use of a wide range of fertilizers), and the lower reaches by intensive urbanization, pollution from industrial processing, and untreated municipal wastewater [3, 17, 33, 34].

Field sampling and laboratory analysis

Samples were collected at sites along the river in Slovenia, Croatia, and Serbia during a high water event in September 2014 and a low water event in September 2015 as part of the GLOBAQUA project [1]. To encompass natural and anthropogenic pollution sources such as industry, transport, and agriculture, 10 and 12 sites were selected in 2014 and 2015, respectively, along the entire course of the Sava River from its headwaters (Sava Bohinjka and Sava Dolinka) to its confluence with the Danube in Belgrade. These sites included Bohinjka (BOH), Mojstrana (MOJ), Radovljica (RAD), Litija (LIT), Vrhovo (VRH) and Čatež (CAT) in Slovenia, Zagreb (ZAG), Jasenovac (JAS), Slavonski Brod (SLB) and Županja (ZUP) in Croatia, and Sremska Mitrovica (SRM), Šabac (SAB) and Belgrade (BEO) in Serbia (Fig. 1, Table S1).

Active acidity and water-soluble salts were determined in the soil and sediment samples at the investigated sites. The pH value of the soil was measured in H_2O using a WTW (Germany) inoLab 7110 pH meter. Water-soluble salts [EC (dS m^{-1})] were measured using a Knick (Germany) Portamess 911 conductometer. To determine the pseudo-total content of the investigated elements, soil samples were collected from the floodplain 10 to 15 m from the river. This zone is most frequently affected by flooding and pollution by heavy metals from the water and river sediments. The soil samples were taken from a depth of 0–10 cm, taking into account that the surface layers of the soil are exposed to the highest accumulations of most heavy metals and metalloids. At each site, soil samples were taken from the top of a 2×2 m^2 area (four samples) and one (fifth sample) from the centre of the square (diagonal intersection) at a depth of 0–10 cm. A composite sample was prepared from these 5 samples. This was done in five replicates (n=5), i.e. in squares, with five composite soil samples obtained for each site. After sampling, the soil samples were transferred to PVC containers and stored in the dark at 4 °C until laboratory analysis. The soil samples

for analysis were air-dried to constant weight, ground in a stainless-steel mill, sieved through a 0.2-mm stainless-steel sieve, and stored in clean polypropylene bags. Each analysis was performed in quintuplicate, and the results were expressed on a dry-weight basis.

The sediment samples were collected using a hand-held benthic landing net. During the expedition in September 2014, only 7 locations were sampled because of flooding and very high water levels. The sampling depth varied depending on the substrate characteristics at each site, typically ranging up to 10 cm in most cases. Wet-sieving was conducted using a coarse 2-mm sieve followed by a 63-μm sieve, with ambient water as the medium. After sieving, samples were transferred to plastic containers and stored in a light-protected environment at a temperature of 4 °C until PTE analysis. All sediment samples were collected according to the recommendations for chemical monitoring of sediments as described in the European Communities Technical Report 2010-041 [35].

To determine the content of PTEs in the soil and sediments, 0.5 g of sample was prepared using the wet digestion method with aqua regia (3 ml HNO_3 and 9 ml HCl) in a microwave oven (CEM Mars 6), according to the USEPA Method 3050B [36] in five independent replicates (n=5). PTE concentrations were determined by optical emission spectrophotometry (ICP-OES, Spectro Genesis) and expressed as mg/kg on a dry-weight basis. The analytical procedure was validated using standard reference material (loam soil–ERM–CC14, IRMM, Institute for Reference Materials and Measurements, Geel, Belgium). As part of the quality control of the laboratory protocol, a standard wet digestion procedure was performed, and the accuracy of the results obtained was in a satisfactory range (100±15%). The detection limits (mg/kg) for the elements studied were as follows: As—0.0051, Cd—0.0002, Cr—0.0010, Cu—0.0007, Ni—0.0003, Pb—0.0042, Zn—0.0062.

Contamination and health risk assessment
Contamination risk assessment

The EF and Igeo were used to determine the extent of contamination levels and the anthropogenic influence on the concentration of the investigated PTEs in soils and sediments. A geochemical methodology that includes the EF and the Igeo is widely accepted to assess the ecological impact of the elements accumulating in soils and sediments [13, 34, 37–39]. The EF was calculated according to the following equation:

$$EF = (Ci/Cr) / (Bi/Br),$$

where C_i/C_r is the ratio between the concentrations of the investigated PTE (C_i) and the concentration of background element (in this case Mn) (C_r) in soil and

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sediments samples; B_i/B_r is the ratio between the referenced background levels in the study area.

The reasons for selecting Mn as a reference element are its predominantly geological origin and limited anthropogenic influence [37, 40]. In addition, principal component analysis (PCA) and strong correlations with the elements studied can help identify a suitable reference background element [3, 41]. In our study, the use of Mn as a reference metal was justified because of its lithogenic origin and its strong correlations with Cr and Ni, both of which are also lithogenic in origin (see Fig. S1). Based on the EF, seven contamination categories are recognized [37]: EF < 1 means no contamination, $1 \leq EF \leq 3$ minor contamination, $3 \leq EF \leq 5$ moderate contamination, $5 \leq EF \leq 10$ moderate to severe contamination, $10 \leq EF \leq 25$ severe contamination, $25 \leq EF \leq 50$ very severe contamination, and $50 \leq EF \leq 100$ extreme contamination.

The Igeo allows comparison between current concentrations of the studied elements and their naturally occurring background concentrations. Igeo was primarily used to determine the contamination level of river sediments [42] but is also commonly used for soils [37, 39]. The Igeo is calculated according to the following equation:

$$Igeo = Ci / (1.5 \times Bi),$$

with Ci the concentration of the studied element in soil and surface sediments samples; Bi the background concentration of the element in soil and surface sediments samples; and 1.5 the correction factor used to eliminate possible variations in background values. Based on the Igeo, five levels of potential ecological risk are recognized [39]: Igeo ≤ 0 low potential risk (I); $0 <$ Igeo ≤ 1 moderate potential risk (II); $1 <$ Igeo ≤ 2 significant potential risk (III); $2 <$ Igeo ≤ 3 high potential risk (IV); and $3 <$ Igeo ≤ 4 very high potential risk (V).

Health risk assessment

An assessment of non-carcinogenic and carcinogenic risks to both children and adults from PTEs was conducted following the guidelines set by the United States Environmental Protection Agency [43]. Three pathways were considered when assessing the health risk from PTEs in soils: ingestion, inhalation, and dermal contact, as described in Baltas et al. [44]. Health risk assessments were conducted independently for each of the sampling sites along the Sava River both during high and low water events. Different coefficients for residents were included in the calculations following USEPA standards [43]. The assessment of non-carcinogenic health risks to children and adults associated with PTE contamination in Sava River sediments focused on ingestion and dermal exposure since these pathways are primary routes of exposure for the population using the water for drinking, agriculture,

and other domestic purposes. The assessment of non-carcinogenic risks associated with PTEs in soils and sediments relied on the calculation of Hazard Quotients (HQs) for each PTE, as described in Jadoon et al. [45] and Varol et al. [21]. However, the evaluation of carcinogenic risk (CRs) was performed only for those PTEs for which specific slope factors were defined.

Non-carcinogenic risk assessment

Following the recommendations, the non-carcinogenic risk for children and adults was calculated using the following equations:

$$HQing = \left[(C \times IRS \times RBA \times EF \times ED) \big/ \right.$$
$$(BW \times AT \times RfDo)(BW \times AT$$
$$\left. \times RfDo) \right] \times 10^{-6},$$

$$HQder = \left[(C \times SA \times AF \times ABSd \times ET \times ED) \big/ \right.$$
$$\left. (BW \times AT \times RfDo \times GIABS) \right] \times 10^{-6},$$

$$HQinh = \left[(C \times EF \times ED) \big/ (AT \times RfC \times PEF) \right].$$

A detailed explanation of the formulas and coefficients used to calculate the individual non-carcinogenic risks is in the Supplementary Material (Table S2 and S3).

The total non-carcinogenic risk for ingestion, inhalation, and dermal contact was calculated using the Hazard Index (HI):

$$HI = HQinh + HQder + HQing.$$

Because the HQs and HI refer to the risks associated with specific elements, their calculated cumulative values were used to provide a more comprehensive representation. This calculation includes the collective effects of all elements studied for each exposure route, referred to as CHQs, and the total effects of all elements across all exposure routes, referred to as CHIs [21]. The cumulative values for HQs and HIs were calculated as the sum of the HQing, HQder, HQinh, and HI values (equations can be found in the Supplementary Material).

Carcinogenic risk assessment

Following the United States Environmental Protection Agency 2020 guidelines [46], the carcinogenic risk assessment for children and adults was conducted using the following equations:

$$CRing = \left[(C \times IFS \times RBA \times CSFo) \big/ AT \right] \times 10^{-6},$$

$$CRder = \left[(C \times DFS \times ABSd \times CSFo) \big/ \right.$$
$$\left. (AT \times GIABS) \right] \times 10^{-6},$$

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$$CRinh = (C \times EF \times ED \times IUR \times 1000) \big/ (AT \times PEF).$$

A detailed explanation of the formulas and coefficients used to calculate the individual carcinogenic risk can be found in the Supplementary Material (Tables S2 and S3).

Assessment of TCR involves summing the individual carcinogenic risks linked to each of the three different exposure routes and is calculated using the following equation:

$$TCR = CRing + CRder + CRinh.$$

The evaluation of the carcinogenic risk resulting from the collective effects of all PTEs studied through the three exposure routes is calculated as the sum of the CRing, CRQder, Crinh, and TCR values (equations can be found in the Supplementary Material).

If the HQ and HI values are less than 1 no negative health effects are expected, and acceptable values for CR range from 10^{-4} to 10^{-6} [44, 46, 47]. Descriptions and reference values for all parameters in the above equations are given in Supplementary Tables S2 and S3. Calculations of HQ, HI, and CR were performed using USEPA's RSL calculator [46].

Statistical analysis

Determination of the relationship between the contents of the studied PTEs in the soil and the surface sediments was performed using Pearson's correlation coefficient. The level of statistical significance is marked with * for $p < 0.05$, ** for $p < 0.01$ and *** for $p < 0.001$. Methods based on PCA and multiple regression analysis (MLRA) were used to determine the origin of chemical elements in the surface layers of soils and sediments. PCA was performed using Varimax rotation and Kaiser normalization; the credibility of the data set was checked using the Kaiser–Meyer–Olkin Measure of Sampling Adequacy (KMO) test and commonalities. The main factors calculated by PCA were then subjected to MLRA of every element, and in this way, the percentage ratio of the factors was determined. The basic statistical analyses, correlation, PCA, and MLRA were carried out using the SPSS Version 21 software package (IBM, Chicago, IL, USA) [48].

Results and discussion

PTE content in riparian soils

The content of PTEs in soils often depends on the type of geological substrate and anthropogenic activities, which can increase their natural background levels several times [49]. Data on the total content of PTE are shown in Fig. 2

and in the Supplementary Material (Table S4). To assess the degree of contamination of the soils studied, we used the natural background of the study area [34] and the natural background for soils in Europe [50] as guidelines.

The concentration of the studied elements during the high (2014) and low water events (2015) in the upper and partly in the middle section of the river was lower than the established guidelines. During 2015, the exceptions to this trend were Pb and Zn, whose values at the MOJ site were above background levels (Fig. 2f and g). The As, Cr, Cu, Ni, and Zn contents in the soils increased from the ZUP site to the BEO site during the high and low water events, indicating potential intensive anthropogenic pressures.
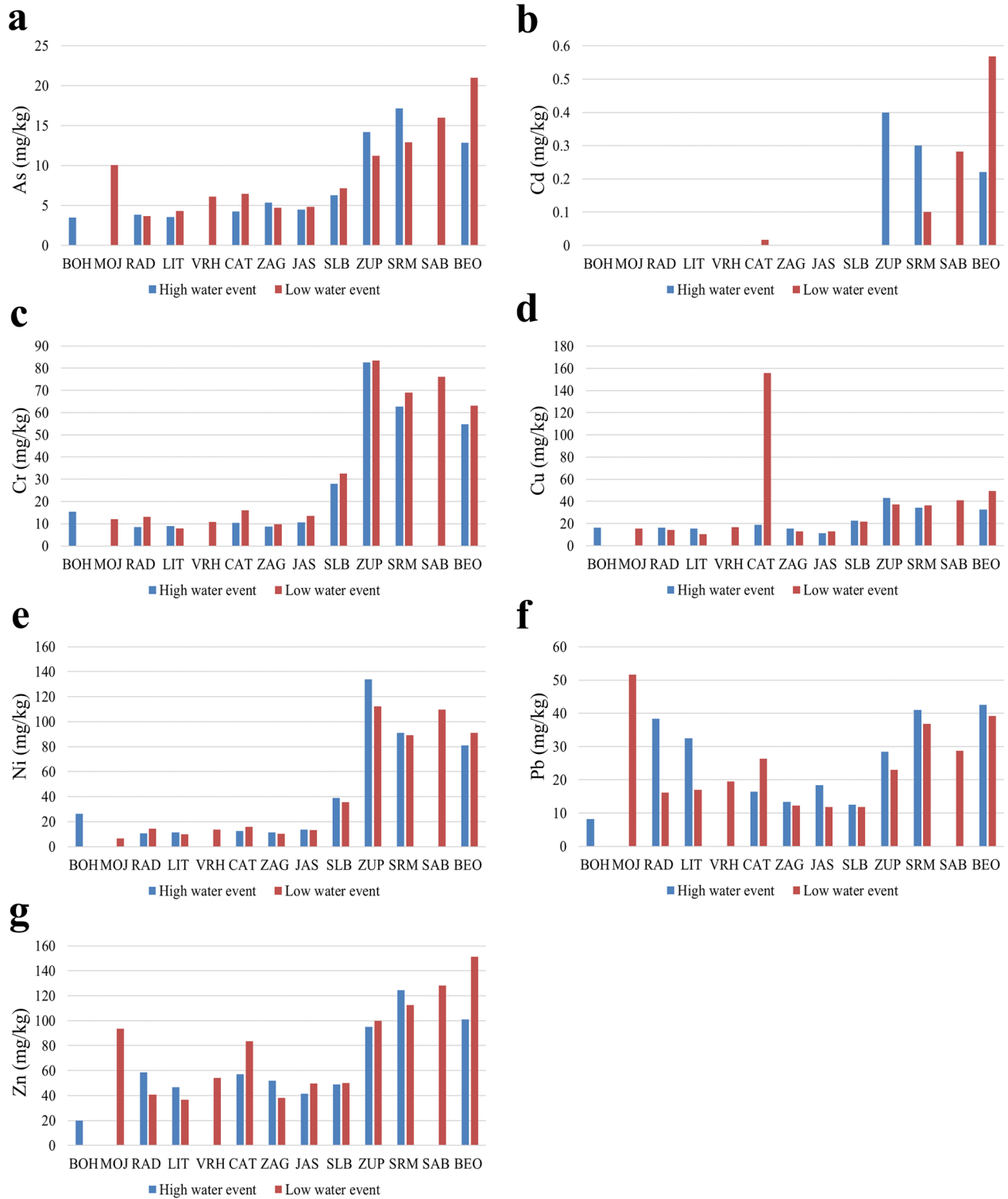
During the high water period, the Pb content increased along the entire river; the contents of other PTEs were lower compared to the low water period (Fig. 2f). The lower PTE concentration during flooding is a consequence of the so-called "dilution effect" [9, 51], as the increased water volume leads to a sudden saturation of the soil and leaching of PTEs into the deeper soil layers.

The bioavailability of elements changes during high water events depending on their properties, changes in pH and oxidation–reduction conditions, the topography of the river, and the duration and type of flood [51, 52]. An increase in pH and EC values often indicates increased mobility of PTEs during flooding [53], which also applies to the soil in the riparian zone of the Sava River during a high water event (Table S4). In our previous studies on the Sava River, we showed that As, Pb, and in part, Ni and Cu are more soluble and available during the high water period [3] than during the low water period [34].
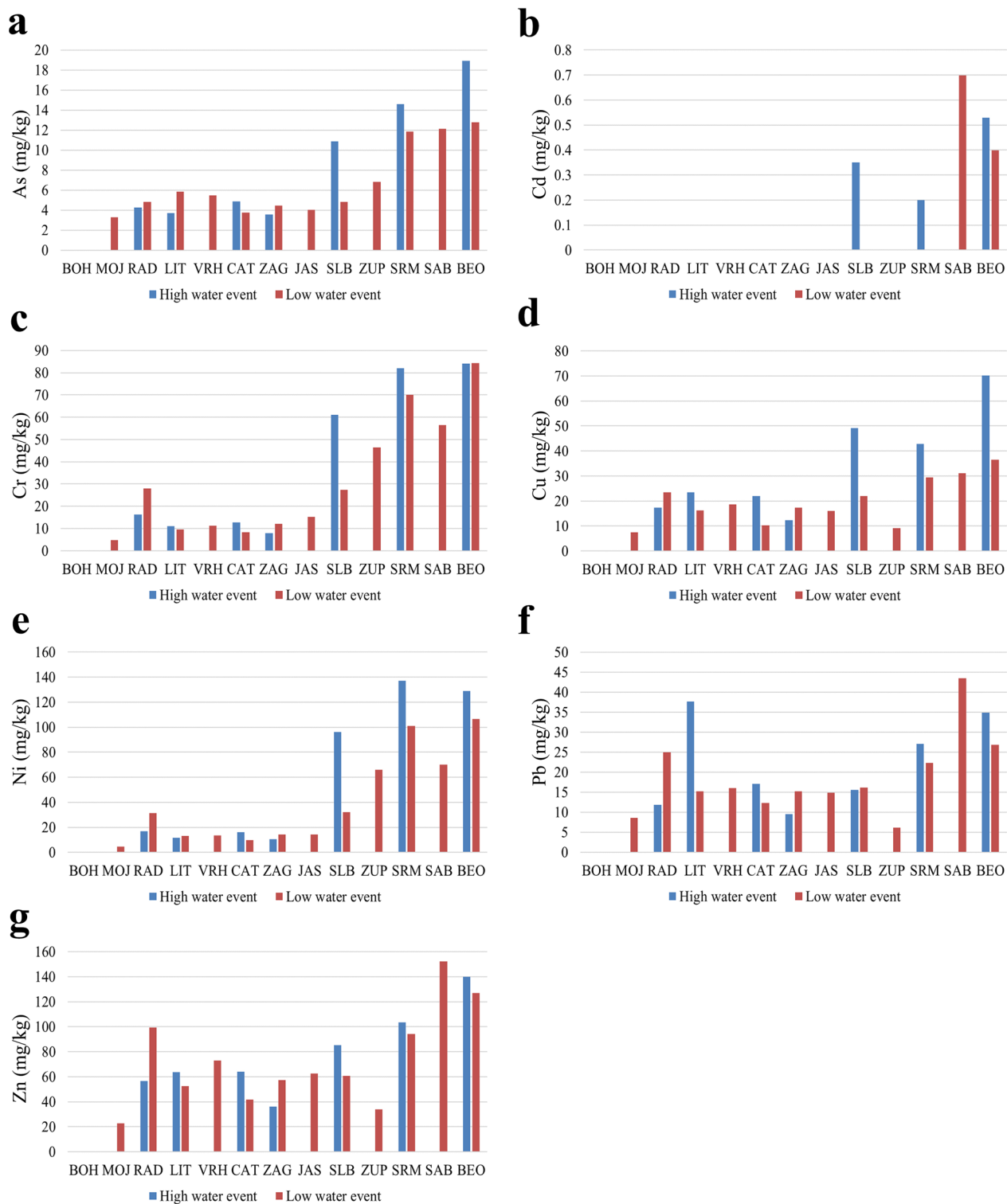
Results show similar concentrations of As, Cd, Cr, Cu, and Zn compared to the soils of the riparian zones of other European rivers with similar anthropogenic pressures (urbanization and industrialization), such as the Elbe [14] and the Danube [54]. The concentrations of Ni and Pb were severalfold higher than in the riparian zone of the Elbe. The concentration of the studied PTEs in the surface layers of the riparian soil was similar to previous studies on the Sava [3, 55].

PTE content in sediments

Data on the total content of PTEs in sediments are shown in Fig. 3 and in the Supplementary Material (Table S5). The Canadian Environmental Quality Guidelines [56] and the natural background of the study area [34] were used to assess the ecological status of sediments. In the Canadian Environmental Quality Guidelines [56], Threshold Effect Limit (TEL) and Probable Effect Limit (PEL) values served to assess sediment pollution. TEL represents threshold levels of PTEs below which no adverse

a

b

c

d

e

f

g

Fig. 2 PTEs in riparian soils during high and low water events

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a

b

c

d

e

f

g

Fig. 3 PTEs in sediments during high and low water events

biological effects are expected, while PEL describes the level of pollutants that affect aquatic life.

In contrast to the soil, the PTE content in the sediments was generally higher during the high water period (Fig. 3 and Table S5). In the upper and partly in the middle sections of the river, the PTE content was below the background values for the riparian zone of the Sava River [26] and below the recommended limits for sediments [56]. The only exception is the RAD site during the low water period, where the content of almost all studied elements was higher (Fig. 3).

An increase in PTEs in the lower section of the river during the high and low water events can also be observed, with all elements studied higher when compared to the TEL values, i.e. above the level of the limiting effect that PTEs can have for the riparian zone and the aquatic community. The Ni content in the lower part of the river was even higher than the PEL value, indicating extreme anthropogenic pollution (Fig. 3e). During extreme flooding as occurred on the Sava River in 2014, large amounts of unpolluted sediments are introduced into the riverbed and the PTE content, especially in suspended solids, decreases [9]. However, even the influx of clean sediments is sometimes insufficient to reduce PTE levels in the lower sections of rivers where pollutant concentrations are highest [51].

After the 2014 extreme high water event, the PTE content decreased. Our results show significantly lower levels of As, Cd, Cr, and Cu in the sediments compared to the concentrations of the same elements in the sediments of larger regional rivers characterized by a similar pollution status, such as the Danube [54]. The Ni and Cr levels were comparable, and those of As, Cd, and Pb in the sediments were several times lower

compared to previous studies in the riparian zone of the Sava [17, 57].

Source identification and contamination risk assessment of PTEs

Based on Pearson's correlation analysis, the studied elements were positively and significantly correlated during the high water event, except for Pb, which indicates that it has a different origin than the other studied PTEs (Fig. S1a). Fig. S1b shows a significant and positive correlation between the studied elements during the low water event, except for Cu and Pb in the soil, which suggests that they might have a predominantly anthropogenic origin compared to the other PTEs.

PCA analysis identified two factors that explained 95.26% of the total variance. The first factor (PC1) explains 84.47% of the total variance and shows the common origin of As, Cd, Cr, Cu, Ni, and Mn, while the second factor (PC2) explains 10.79% of the total variance and indicates a strong anthropogenic influence on the origin of Pb and partly of Zn in soil and sediments during the high water event (Fig. 4a).

Analysis revealed two distinct factors that accounted for 81.70% of the total variance, as shown in Table S6. PC1 explained 67.34% of the total variance and indicated a predominantly geological origin of Ni, Cr, Mn, and As, and their influence is mainly observed in the middle and lower sections of the river. The second component explained 14.36% of the total variance and indicated significant anthropogenic inputs of Cu to soils, as well as Pb and Zn to soils and sediments during the low water event (Fig. 4b); Cd was observed in both the 1st and 2nd clusters with equally significant bindings, indicating its dual origin influenced by both geologic and anthropogenic factors, depending on the particular sampling site.
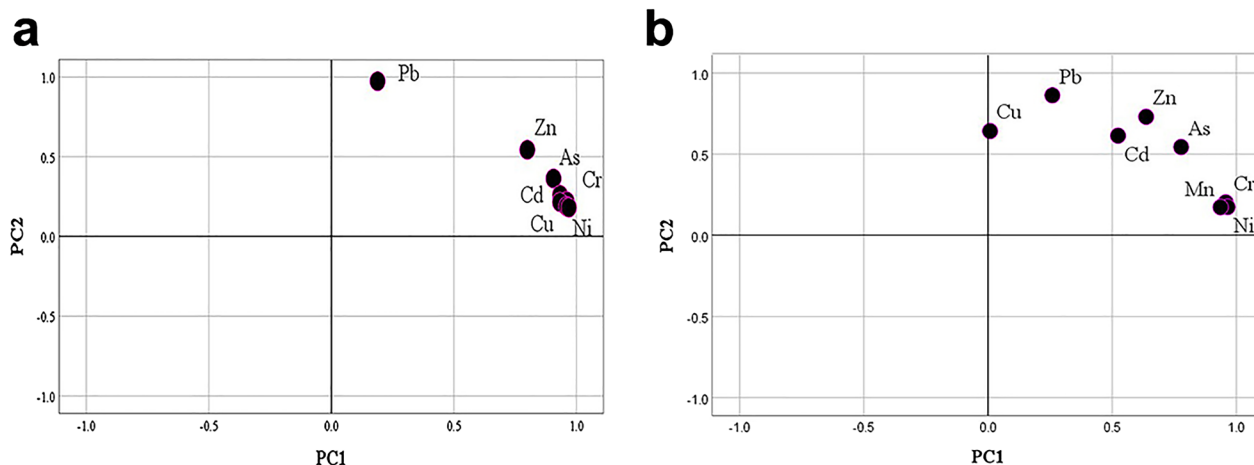
Fig. 4 PCA for soil and sediment samples in the Sava River riparian zone during high and low water events: **a** high water event, **b** low water event

Based on the average EF values in the soils during the high water event, there was slight enrichment with As (1.03), Cu (1.60), Ni (1.38), Pb (1.05), and Zn (1.22) (Table S7). The average EF values in the soils during the low water event showed minor enrichment for As (1.21), Cu (2.82), Ni (1.46), and Zn (1.34). Exceptions were As (3.58), Pb (4.81), and Zn (4.18), for which moderate enrichment was observed at MOJ, and Cu (14.16), where significant enrichment was observed at the CAT site (Table S7).

During the high water event, EF values in the sediments showed a minor enrichment with Cu (1.95), Ni (1.47), and Zn (1.33), except at the LIT site, where a moderate enrichment with Cu (3.07) was observed (Table S9). The average EF values during the low water period showed minor enrichment with As (1.02), Cu (1.51), Ni (1.53), and Zn (1.51) (Table S9).

Although the results reveal minor contamination by the investigated PTEs, EF values > 1.5 for As, Cu, Ni, Pb, and Zn indicate their anthropogenic origin and high potential contamination risk.

Based on the average Igeo values in the soils during the high water event, most sites along the river were at low potential contamination risk, except Ni at the ZUP, SRM, and BEO sites (1.11, 0.55, and 0.39, respectively), where a moderate potential risk was identified (Table S8). The average Igeo values for the soils during the low water period also indicated a low potential risk for As, Cd, Cr, Pb, and Zn. The average Igeo values indicated a moderate contamination risk for Cu and Ni in the lower part of the river and a high contamination risk for Cu (2.10) at the CAT site (Table S8).

Similar to the soil samples, the average Igeo values in the sediments indicated a low potential contamination risk for As, Cd, Cr, Pb, and Zn under both high and low water events (Table S10). Exceptions were Cu and Ni at the SLB, SRM, and BEO sites, with values between 0.24 and 1.06, indicating moderate to significant potential contamination risk (Table S10).

PCA showed a mixed origin for As in soils and sediments, indicating a predominantly geological origin with the influence of anthropogenic factors, especially in the lower section of the river, which was also confirmed by the MLRA analysis (Fig. S2). The low As content and the negative values of Igeo during both high and low water events (Tables S8 and S10) indicate that the origin of As in the upper and middle sections of the river is predominantly geological, resulting from the decomposition process of the parent material. The increase in As in the lower section of the river indicates accumulation mostly from anthropogenic diffuse sources such as industrial and agricultural activities [3, 34], as well as road and railroad traffic [58]. There are two oxidation states of arsenic

in nature, As(V) and As(III), with the trivalent form being significantly more toxic to the environment with increasing pH [59]. The increase in As content in the lower section of the river (Tables S4 and S5), the redox condition, and the anthropogenic influence on its origin indicate a possible change to As(III), as also shown by the Igeo and EF (Tables S7 to S10). According to PCA, the change occurred in both clusters with equally strong connections during both research periods. This indicates that its origin is influenced by both geological and anthropogenic factors (Fig. 4), which was also confirmed by MLRA (Fig. S2). The Cd in the upper section of the Sava River is predominantly of geological origin, from ore deposits and a specific geological substrate [17, 60]. The origin of the Cd appears to be partly due to historical Zn and Pb mining [61] and industrial activities associated with Zn smelting in locations upstream of the CAT site [17], with the effects being transported downstream. Cd levels in the lower section of the river are mostly above background levels due to intensive agricultural production and the use of phosphate fertilizers [62], the discharge of untreated industrial and municipal wastewater [32], and intensive atmospheric emissions originating from thermal power plants [63]. Cd occurs in soil as a divalent cation (Cd^{2+}) at concentrations typically between 0.1 and 1.0 mg/kg and is rarely found in pure form in soil. It occurs more frequently in combination with Zn, Cu, and Pb. Its mobility and availability in the soil increase with increasing pH [59], confirmed by earlier studies of the Sava River [3, 34]. Due to its high solubility and availability, Cd can pose a significant contamination risk, especially in the lower section of the river, as indicated by the EF values (Tables S7 and S9).

Cr and Ni are grouped in the first cluster, indicating their predominant geological origin in both research periods, which was also confirmed by MLRA (Fig. S2). The influence of the central Dinaric ophiolitic belt is noticeable at the ZUP site, characterized by Cr-rich serpentinized rocks. In this part of the river, the increased Cr and Ni contents are due to the decomposition of the bedrock and the subsequent leaching of soil particles by alluvial and erosion processes in the Bosna [58, 64] and Kolubara catchments [13, 65]. Anthropogenic activities in the lower part of the river increase the Cr content in the soil due to discharges of untreated or partially treated industrial wastewater from smelters and steelworks between SLB and SAB [17]. Agricultural activities also contribute to the anthropogenic origin of Cr as a result the excessive use of organic livestock and poultry manure [66], which is confirmed by the indices (EF and Igeo), indicating low contamination risk (Tables S7–S10). At the SLB site, the elevated Ni content is attributed to its greater mobility compared to Cr [34, 58] and anthropogenic activities

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such as forging and wood processing plants [17], with the EF and Igeo indicating a medium to significant contamination risk to soil and sediments (Tables S7 to S10). Cr in the soil can be present in different oxidation states (+2 to+6), with hexavalent chromate Cr(VI) and trivalent chromite Cr(III) being the most important from an environmental perspective as the most stable and dominant forms of Cr in nature. The speciation of Cr depends on factors such as redox potential and pH reaction [67]. Cr(VI) is considered in the assessment of environmental and health risks. In neutral and slightly alkaline soils, Ni occurs in the form of Ni(II) hydroxide with low mobility and solubility [59]. Its extremely high content in the lower section (Tables S4 and S5) and its anthropogenic origin point to a high risk of contamination.

During the low water event, Cu is singled out in the 2nd cluster, indicating its strong association with anthropogenic factors (Fig. 4b). Also, as Cu is not correlated with any other element in the soil, its origin is specific, and a consequence of anthropogenic diffuse pollution (Fig. S1a). The highest levels of Cu occur in the middle and lower sections of the Sava (Figs. 2 and 3). Considering that the middle section of the river (CAT, ZAG, SLB, and JAS) is part of wine-growing regions, the increased Cu content in the soil could be due to the use of pesticides and herbicides in viticulture [68]. The increased Cu content at the localities in the lower part of the river is partly the result of the transfer of influences from the upstream localities and the result of intensive industrial and agricultural activities [3, 69], as well as the discharge of industrial and municipal wastewaters [60]. The EF and Igeo showed that Cu contamination of soils and sediments along the entire course of the Sava River is moderate, except at the CAT site during the low water event, where there is significant contamination (Tables S7–S10). Although Cu occurs naturally as Cu (I) and Cu (II), it is most commonly found as Cu (II) in aerated surface soils, and it is this form that is associated with its bioavailability and toxicity [70]. Because of elevated EF and Igeo values and the anthropogenic source of Cu in both soil and sediments, it poses a significant potential contamination risk to the environment. Pb was assigned to the 2nd cluster in both research periods, indicating a predominantly anthropogenic origin, confirmed by the MLRA (Fig. S2). The elevated Pb concentration in the upper section of the river could originate from point sources such as ore deposits and is related to mining and intensive industrial activities, i.e. metal processing industries upstream of the studied site [60], with EF indicating moderate potential pollution (Tables S7 and S9). A slight increase in Pb content was observed in the lower section, and its anthropogenic origin could be explained by industrial activities, predominantly diffuse pollution sources such as railroad

traffic, and the emission of harmful gases from motor vehicles [73]. The pH value has a major influence on the mobility and solubility of Pb in soils and sediments, as at low pH its solubility is increased, whereas at pH>8, it precipitates in the form of hydroxide, phosphate, or carbonate, which makes it extremely stable and unavailable [70]. This is also confirmed by the results of previous studies on the Sava River at low water levels [34]. EF shows that the entire riparian zone is slightly contaminated with Pb (Tables S7 and S9), confirming its stability and low availability.

The anthropogenic factor has the greatest influence on the origin of Zn in soil and sediments, as PCA and MLRA show (Fig. S2). In the upper part of the river, the origin of Zn is similar to that of Pb, i.e. the increased content is the result of mining activities, which confirms its mixed origin [34]. Downstream, a trend of increasing Zn content in the soil was observed, which is particularly evident near large settlements (ZAG, BEO) and near industrial centres (SAB, SRM) (Fig. 4). In the middle section of the river, Zn in the soil originates from agricultural activities, such as the use of different pesticides and fertilizers, but also from the discharge of industrial and untreated or partially treated municipal water [17]. Agricultural and industrial activities [34] have a significant impact, suggesting that the origin of Zn in the lower section of the river during high and low water events is anthropogenic. Like Cd, Zn occurs most frequently in a bivalent form (Zn^{2+}). The solubility of Zn is highest in an acidic environment at a very low pH, and as the pH increases, especially above 6.5, Zn tends to form highly stable complexes that are nearly unavailable to biota [59]. Despite its low availability, the EF shows that contamination is moderate along the entire length of the river, in particular in the upper section of the river, i.e. the MOJ site, where pollution is significant during a low water event. The Igeo indicates a moderate contamination risk in the lower section of the river during high and low water events (Tables S7 to S10).

Potential human health risk assessment
Non-carcinogenic risk assessment of PTEs in soil
In the SRB region, riparian zones are frequently used as agricultural lands and orchards, often irrigated by the rivers. Riparian zones and rivers are also frequently used for various recreational activities. Therefore, it is important to monitor PTE pollution and evaluate its impact on human health, especially during and after flooding, as PTEs can travel over long distances and cause major socio-economic and health problems for the population in riparian areas. Hence, assessing health risks at each sampling site can identify risk zones where adults and children are exposed to pollutants [71].

The individual non-carcinogenic risk values from ingestion for children were mostly below 1 and acceptable at all sampling sites; however, cumulative values (CHQing) exceeded 1 in the lower section of the river during high- and low water events, indicating a significant non-carcinogenic risk associated with the ingestion of PTEs in the soils studied. In contrast to ingestion, the cumulative values for dermal uptake and inhalation were consistently below 1, indicating that the presence of PTEs in the soil does not pose a significant
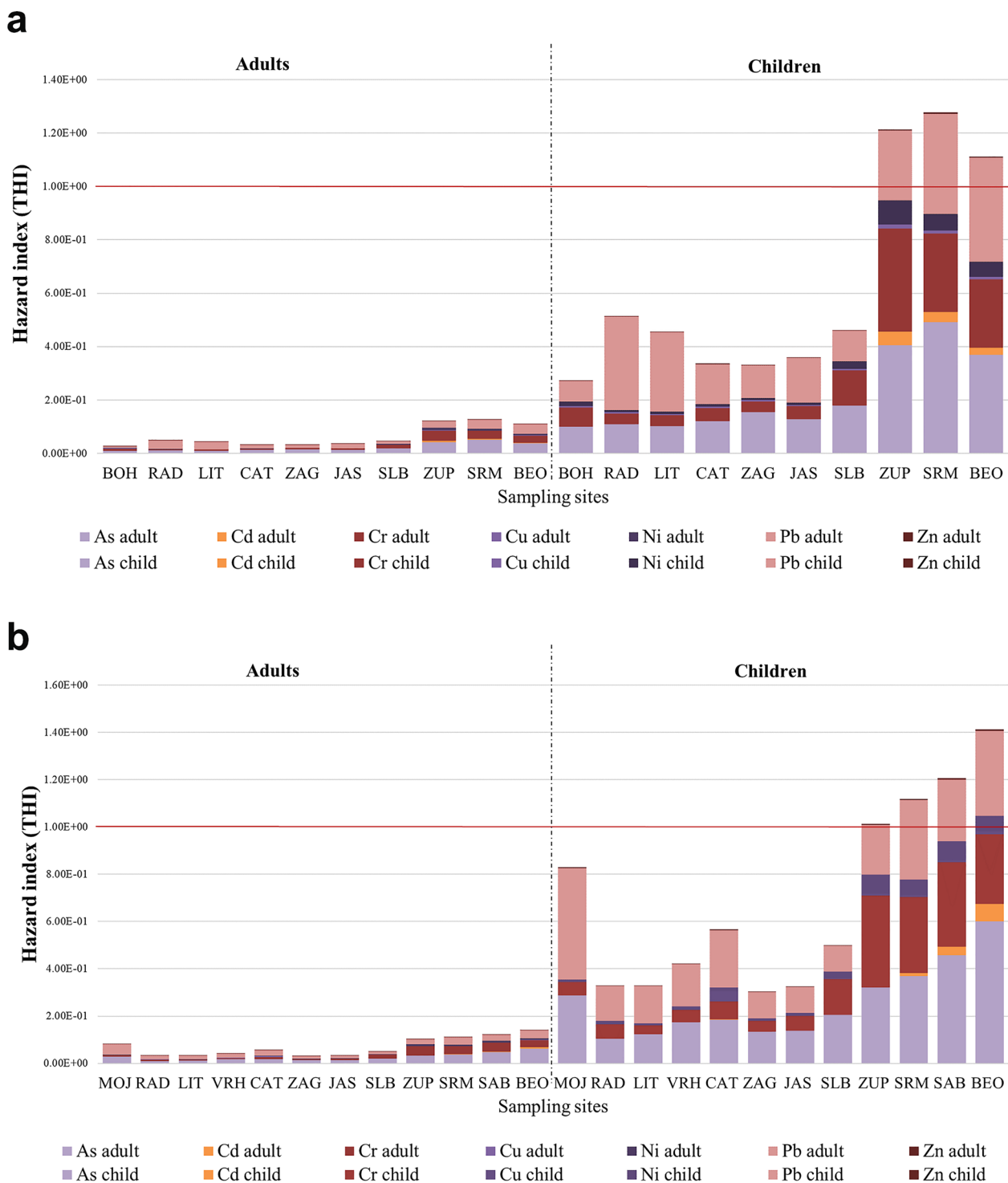
Fig. 5 Non-carcinogenic risk to adults and children of PTEs in soils: **a** high water event, **b** low water event

non-carcinogenic risk when these routes of exposure are considered (Table S11). The cumulative hazard quotient for adults via all exposure routes was consistently below 1 at all the sites, and the cumulative THI for adults also remained below the threshold (Fig. 5), indicating that there is no non-carcinogenic risk from the PTEs in soils during high and low water events (Table S12).

The cumulative THI levels for children in downstream areas exceed the threshold value of 1, with ingestion being the greatest contributor to overall non-carcinogenic risk. Similar conclusions were reached by other researchers when considering the effects of PTEs on human health [72–74]. The highest THI value was observed for As, followed by Pb, Cr, and to some extent Cd (Fig. 5). This is consistent with previous findings where it was concluded that As poses the greatest non-carcinogenic risk [75, 76].

These results show that the residents of the Sava River basin are most susceptible to As ingestion compared to Pb, Cr, and Cd (Fig. 5). When comparing the HI values for children and adults, the values for children are significantly higher, underscoring their increased sensitivity to the adverse effects of PTEs due to their lower body mass and specific hygiene habits. Studies have consistently found elevated THI levels in children [19, 44, 72, 74, 76].

Considering the high concentrations of As, Cd, Cr, and Pb in the lower section of the river, the potential contamination risk based on EF and Igeo, and their bioavailability and solubility [3, 34], it can be concluded that they pose a high risk to the health of children, but also adults, regardless of the low cumulative THI values.
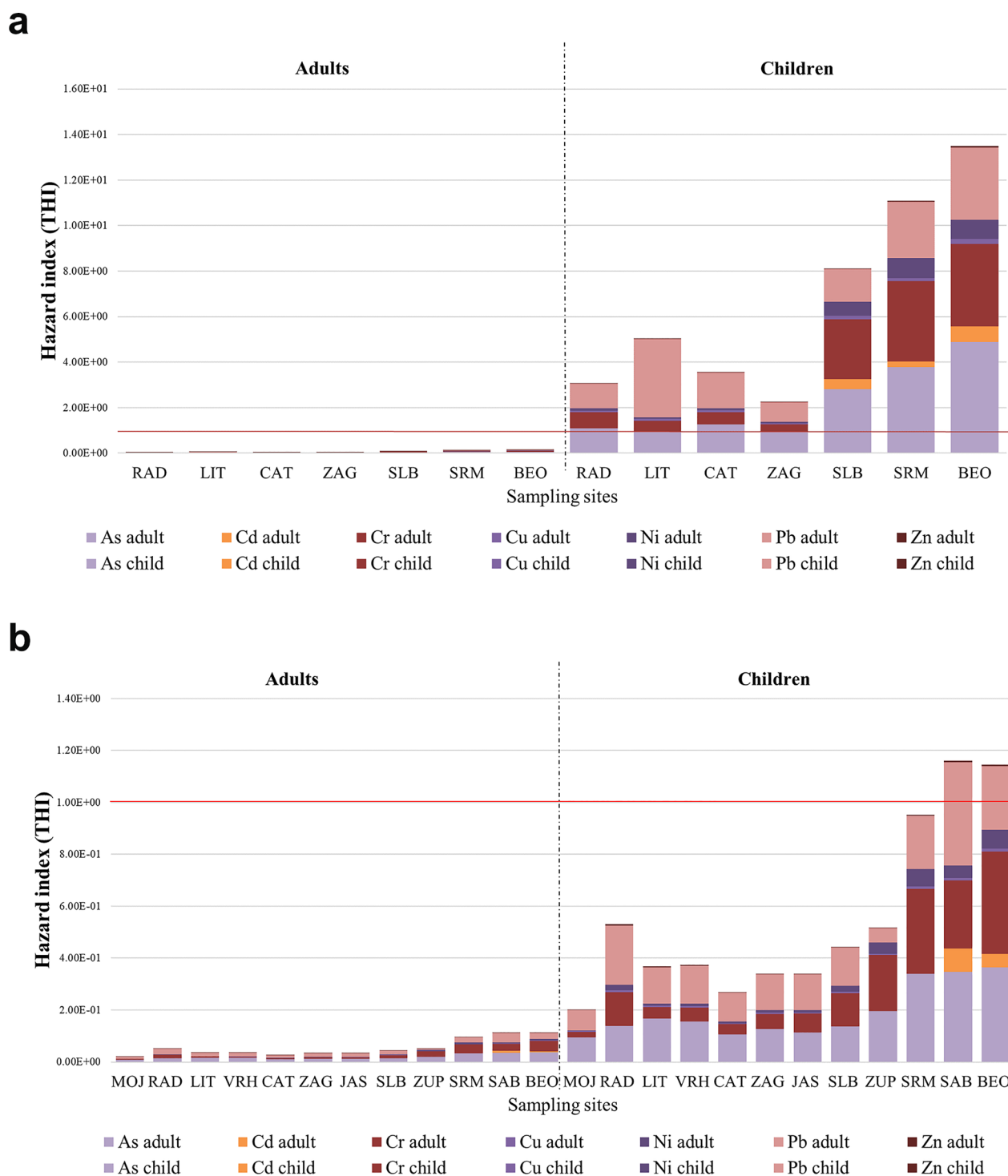
Carcinogenic risk assessment of PTEs in soils

The cumulative carcinogenic risk values for As, Cr, Pb, and the TCR values were all within the acceptable range (10^{-4} to 10^{-6}) as defined by the US Environmental Protection Agency [46]. These results indicate that no carcinogenic risks were expected for residential areas. Detailed results are provided in Table S13.

TCR values decreased in the following order: CRing > CRder > CRinh, a pattern corroborated by previous studies [21]. Ingestion values contributed to 88% of the total cumulative TCR value, significantly exceeding both dermal and inhalation carcinogenic risk values severalfold. Regarding the geographical distribution of carcinogenic risk by sampling site, it was found that despite the risk being classified as "acceptable", the inhabitants of the lower part of the river (from SLB to BEO site) are potentially exposed to a higher risk given the high levels of As, Cr, and Pb, their availability, and the potential contamination risk based on the EF and Igeo. Cr was also found to be the largest contributor to TCR via ingestion, dermal contact, and inhalation, followed by As and Pb (Fig. 6). These results are consistent with the findings of Varol et al. [21] and Cai et al. [74].
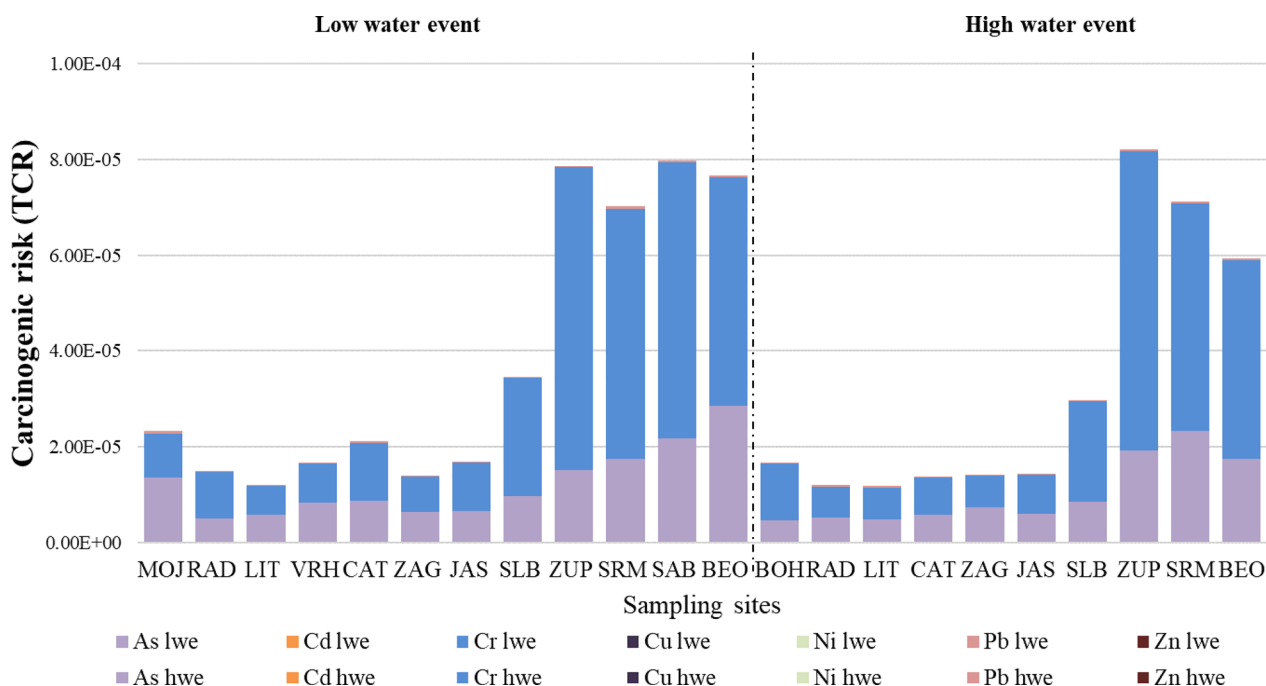
Fig. 6 Carcinogenic risk to residents of the Sava River riparian zone of PTEs in soils. Hwe—high water event; lwe—low water event

a

b

Fig. 7 Non-carcinogenic risk to adults and children of PTEs in sediments: **a** high water event, **b** low water event

Non-carcinogenic risk assessment from PTEs in sediments

During the low water period, HQing values for children were < 1 for all elements, but cumulative ingestion values in the lower section of the river were higher than the threshold (< 1; 46), suggesting that all PTEs in sediments pose a significant risk to children via the ingestion route (Fig. 7b). During the high water period, HQing levels for As, Cr, and Pb for children were several times higher than
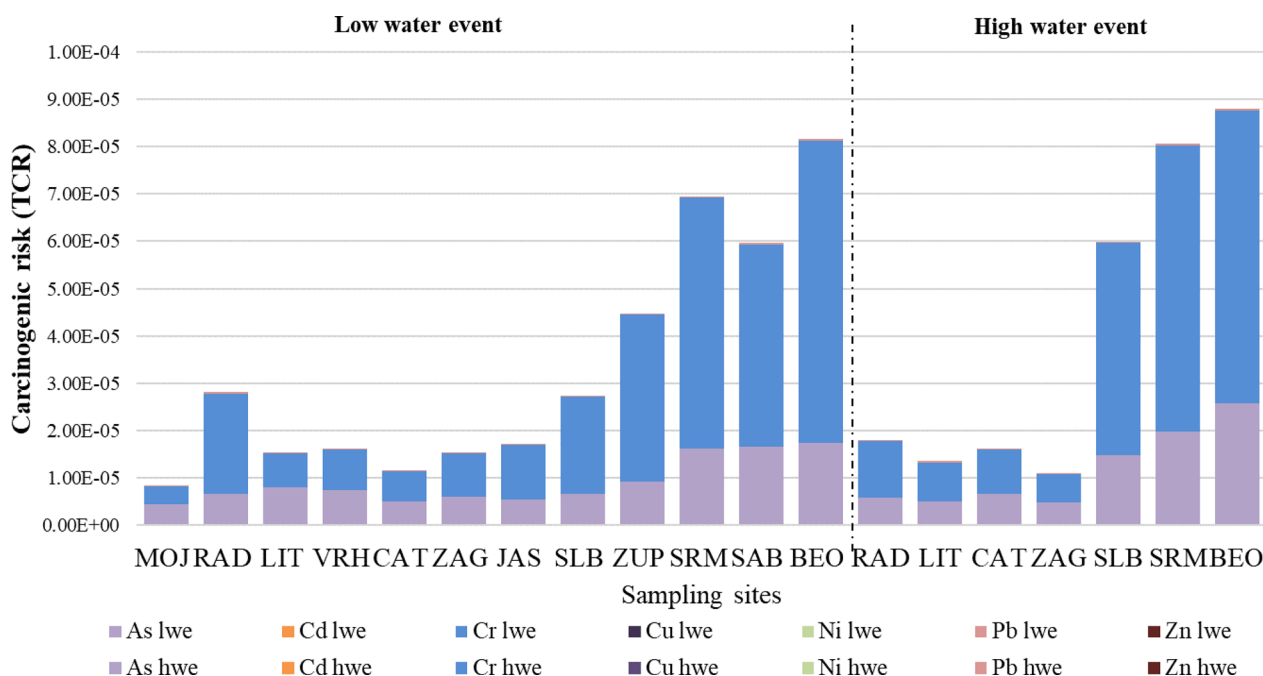
Fig. 8 Carcinogenic risk to residents of the Sava River riparian zone of PTEs in sediments. Hwe—high water event; lwe—low water event

the threshold (< 1), with cumulative levels 10 times higher than recommended, indicating an exceptional non-carcinogenic threat from these metals in sediments (Fig. 7a). Values for both individual and cumulative dermal exposure risk were < 1 during both high and low water events, supporting the conclusion that the PTEs present in the sediments do not pose a significant non-carcinogenic risk when the dermal exposure route is considered (Fig. 7; Table S14). Ingestion of PTEs poses a higher risk to children, which is consistent with previous studies [77, 78].

Cumulative health values for adults via all routes of exposure were below the threshold (< 1) at all sites, indicating that there is no significant non-carcinogenic risk from PTEs in sediments during high and low water conditions in the riparian zone of the Sava River (Fig. 7; Table S15).

As with soils, cumulative THI values for children in the downstream area exceed the threshold of 1 several times over during both high and low water events, with ingestion being the primary route contributing to the overall non-carcinogenic risk (Fig. 7). Similar conclusions were reached when the impact of PTEs on human health was considered [77–79]. The sediment analysis indicates that SRB residents are most at risk via the ingestion route, with As, Cd, Cr, and Pb posing the greatest risk. Overall health risk assessments for children and adults revealed significantly higher values for children, underscoring their heightened vulnerability to the adverse health effects of sediment-borne PTEs compared to adults.

Carcinogenic risk assessment from PTEs in sediments

The cumulative carcinogenic risk values for As, Cr, and Pb from ingestion and dermal exposure, as well as the total carcinogenic risk values, were all within the acceptable range of 10^{-4}–10^{-6} as defined by the USEPA [46]. This indicates that no carcinogenic risks were expected for the residents (Fig. 8). Detailed results can be found in Table S13.

As part of the carcinogenic risk assessment by sampling site, analysis showed that residents of the lower Sava River section, which extends from SLB to BEO, and residents of the RAD site, are at higher carcinogenic risk given the high levels of As and Cr and the potential contamination risk based on the EF and Igeo.

Conclusions

This study utilized various methods, guidelines, and health and contamination risk assessment indices to evaluate soil and sediment contamination and its impact on human health in the riparian zone of the Sava River during high and low water events. The contents of As, Cr, Cu, Ni, and Zn in the soil in the lower section of the river (ZUP to BEO) were higher than the background values for the riparian zone of the Sava River during the two periods. The Ni content in the lower section of the river and the Cu content at the CAT site were higher than the background values for European soils, indicating a potentially high risk of contamination. According to the SQGs, the As, Cd, and Cr concentrations in the

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sediments in the lower part of the river and the contents of Cu, Ni, and Zn in the lower and upper sections of the river were above the proposed TEL values. In the lower section of the river from the ZUP to BEO, the Ni content was up to threefold higher than the PEL values, indicating high contamination and potential risk. The results for the EF and Igeo indicate that the potential contamination risk is significantly higher in the lower section of the river where an anthropogenic impact is observable and that As, Cu, Ni, and Zn contribute the most to the contamination risk. PCA and MLRA showed that Cr is mainly of geological origin and has a limited impact on contamination risk. As and Ni have geological and anthropogenic origins, with anthropogenic influences notable in the lower section of the river. Cu, Pb, and Zn are primarily of anthropogenic origin and contribute to a moderate to high contamination risk in the lower part of the study area. This was also confirmed by the EF and Igeo assessments. The results of the health risk assessment for soils and sediments showed that ingestion is the primary route of exposure for adults and children and that As, Cd, Cr, and Pb potentially pose the greatest non-carcinogenic risk, with children at higher risk, in the lower section of the river. The values for carcinogenic risk were within acceptable limits. Given the elevated levels of As, Cd, Cr, and Pb in the lower section of the river, it can be inferred that these elements pose a significant contamination risk based on the EF, Igeo, and their bioavailability and solubility. This represents a high potential health risk to the population of the SRB, despite the low cumulative THI and TCR values.

Insights into potential health risks and associated contamination with PTEs are crucial when planning future activities and sustainable provision of ecosystem services for riparian zones of large river basins globally, including sustainable development and preservation of riparian areas. This includes the collection and systematization of all available data on anthropogenic pressures, contamination levels, and associated environmental and health risks. At the same time, this research can provide a foundation for implementing measures and recommendations that contribute to the harmonization and improvement of environmental and health policy by scientific findings. This primarily entails establishing a comprehensive network of monitoring stations to oversee the quality of riparian soil and sediments and ensuring public access to the collected data, especially considering the importance of the Sava River as a major regional watercourse and primary source of drinking water for a large number of inhabitants.

Based on all these data, it is possible to identify potential risk zones where adults, and children in particular, are exposed to non-carcinogenic and carcinogenic risks associated with PTEs. This information can guide the development of pollution mitigation plans and sustainable ecosystem services in riparian zones.

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1186/s12302-024-00952-3.

Additional file 1.

Declarations

Ethics approval and consent to participate
Not applicable.

Consent for publication
All authors have read and approved the final version of the manuscript. All authors have given their consent to the publication of the manuscript.

Competing interests
The authors declare that they have no competing interests.

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