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From a single waste source to secondary diffuse pollution: heavy metal contamination of sequential floodplains triggered by coal waste stockpiling

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Abstract. Floodplains represent key depositional environments within river systems, acting as long-term sinks for sediments and associated contaminants transported during flood events. This study investigates the spatial distribution and accumulation of heavy metals across a sequence of ten floodplains within a defined river segment downstream of a single contamination source — an abandoned coal-waste stockpile in the Krraba tributary of the Erzen river, central Albania. Sediment and soil samples (n = 30) were analysed for Cd, Pb, Cr and Cu by flame atomic absorption spectrometry after HF/$HClO_4$ digestion. Pollution levels were assessed using CF, I_{geo}, PLI and EF, referenced to the FOREGS European topsoil baseline. All floodplains are classified as extremely contaminated, with PLI values (n = 4) ranging from 40.6 to 53.4. Cadmium shows the highest enrichment (CF 400–597; I_{geo} 8.0–8.6; EF 30–62), followed by Pb and Cu, while Cr levels reflect a persistent ultramafic signature transported from the upper catchment sediments. By analyzing a connected sequence of floodplains within a single contaminated river segment rather than isolated units, the study reveals the spatial evolution of contamination along a downstream gradient and demonstrates how a local point source generates a persistent secondary pollution footprint, with practical implications for floodplain monitoring and river-basin management.

1 Introduction

River systems are fundamental for transporting sediments and associated contaminants, particularly heavy metals, which are persistent and toxic in downstream ecosystems [1]. While water quality fluctuates, sediment-bound contaminants often persist for decades, representing a long-term environmental risk. Floodplains function as natural depositional zones during flood events, where fine-grained sediments enriched with organic matter trap heavy metals through adsorption [2]. This process transforms floodplains into long-term sinks that record historical contamination, often extending pollution well beyond the original

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source [3]. Similar integrated approaches combining geochemical indices and sedimentological controls have been widely applied in river contamination studies [4-6].

In many basins, a single point source — such as industrial or mining waste — can generate a sequence of contaminated floodplains downstream through repeated flooding [7, 8]. Despite their potential impact on agriculture and human health, most studies focus on riverbed sediments or isolated sites rather than on the cumulative nature of floodplain contamination. In Albania, these environments remain largely outside systematic monitoring and rehabilitation frameworks.

The present study addresses this gap by investigating the spatial evolution of heavy metals (Cd, Pb, Cr, Cu) across a sequence of ten floodplains downstream of an abandoned coal-waste stockpile in the Erzen river. Specifically, the research aims to (i) quantify heavy-metal concentrations using flame atomic absorption spectrometry, (ii) evaluate spatial trends along a downstream gradient using multiple pollution indices (CF, I_{geo}, PLI and EF), and (iii) demonstrate how floodplains transform localised contamination into a persistent secondary pollution footprint, in alignment with the EU Water Framework Directive [9].

2 Study area

The study area is part of the Erzen river basin in central Albania, extending from the confluence of the Krraba stream with the main Erzen channel downstream to the floodplain near the Qehas bridge (Figure 1). The coal-waste stockpile, which constitutes the primary source of pollution, is located within the Krraba tributary sub-catchment. These wastes were generated during mining activities in the Krrabë–Mushqeta area between 1968 and 1995. After the closure of the mine, the initial volume was approximately 219,252 m³, whereas today only a much smaller amount remains, around 10,000 m³. The Erzen basin covers 760 km² with a channel length of 109 km; its main tributaries are the Krraba (79.8 km²), Zhullimë (132 km²) and Peza (74.3 km²) streams.

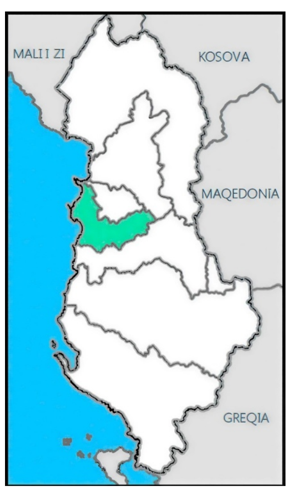

Fig. 1. Location of the Erzen river basin in central Albania and position of the study reach on the Krraba tributary.

2.1 Climate and hydrology

The basin receives an average annual rainfall of 1248 mm. At the downstream Sallmonaj station the mean annual discharge (1960–2008) is 18.3 m³ s⁻¹. For the Krraba tributary at the

Ibë station, flow characteristics include a maximum discharge of 3.50 $m^3\ s^{-1}$, with Q50 = 2.3 $m^3\ s^{-1}$ and Q90 = 1.09 $m^3\ s^{-1}$. Extreme floods occur during the 10% of time corresponding to peak discharges, while frequent floods are of moderate magnitude.

2.2 Floodplains under investigation

Ten floodplains (FP1–FP10) were selected along the study reach. Individual floodplain surface areas range from 7 300 to 10 300 m^2, and their mean distance from the river axis ranges from 32 to 60 m. The sediments exhibit a heterogeneous alluvial texture, consisting of coarse fractions (gravel and sand) interbedded with finer materials (silt and clay). The deposits reflect variable hydrodynamic conditions associated with flood events, resulting in poorly sorted sediments with low organic-matter content. Coal-derived fragments are visible at all floodplain sites (Figure 2). Land use along the investigated reach is predominantly natural; anthropogenic interventions are limited to two bridges whose support columns lie adjacent to the studied floodplains.

a) b) c)

Fig. 2. Field photographs showing coal-derived fragments at representative floodplain sites: (a) the most downstream (tenth) floodplain; (b) an intermediate floodplain; and (c) the floodplain located adjacent to the coal waste stockpile. (photo: M. Hoxhaj, 2025).

3 Materials and methods

3.1 Sampling strategy

Sediment and soil samples (n = 30) were collected from ten floodplain sites at increasing distances downstream of the pollution source. At each site, three replicate sub-samples were composited to account for spatial variability. Surface sediments (0–10 cm) were taken from riverbanks, floodplain centres and distal margins, while deeper samples (down to 30 cm, below the gravel cover) were retrieved to evaluate historical deposits (Figure 3). Sampling was carried out during the post-flood period (May–June 2025); tools were cleaned between sites, and samples were stored in polyethylene bags at 4 °C prior to analysis [2].

3.2 Laboratory analysis

Dried and sieved sub-samples were digested in platinum crucibles with HF and $HClO_4$, transferred to 100 mL volumetric flasks with distilled water, and analysed for Cd, Pb, Cr and Cu by flame atomic absorption spectrometry (FAAS). Analytical quality was controlled through procedural blanks and duplicate analyses; recoveries were within ±10% of the expected values [2].

Fig. 3. Map of the sequential floodplains (FP1–FP10) along the Erzen river, downstream of the coal-waste stockpile on the Krraba tributary.

3.3 Pollution indices and data analysis

Pollution levels were assessed using the Contamination Factor (CF), the Geoaccumulation Index (I_{geo}), the Pollution Load Index (PLI) and the Enrichment Factor (EF). The geochemical baseline adopted throughout the study is the median European topsoil (0–25 cm) composition from the FOREGS geochemical atlas [10]: Cd 0.145, Pb 15, Cr 22, Cu 13 mg kg^{-1}. This harmonised European reference is preferred to the world-shale composition because it is the recommended benchmark for soil-contamination assessments under EU regulatory frameworks [9, 11].

Contamination Factor [12]:

$$CF_i = C_i / C_{background} \tag{1}$$

Geoaccumulation Index [13]:

$$I_{geo} = \log_2 [\, C_i / (1.5 \cdot C_{background}) \,] \tag{2}$$

where the factor 1.5 accounts for natural variability of the background due to lithogenic effects [13].

Pollution Load Index [14]:

$$PLI = (CF_1 \cdot CF_2 \cdot \ldots \cdot CF_n)^{(1/n)} \tag{3}$$

with n = 4 metals (Cd, Pb, Cr, Cu).

Enrichment Factor [12]:

$$EF = (C_i / C_{ref})_{sample} / (C_i / C_{ref})_{background} \tag{4}$$

Chromium (Cr) was used as a reference element for Enrichment Factor (EF) calculations due to its common application in sediment geochemistry. However, the upper Erzen River basin is influenced by ophiolitic formations of the Albanian ultramafic belt, characterized by Cr-rich lithologies such as chromite-bearing peridotites [15]. This lithological background may cause natural variability in Cr concentrations, particularly in upstream areas, introducing some uncertainty in assuming Cr as a fully conservative element for EF interpretation. Classification thresholds follow Müller [13] for I_{geo}, Håkanson [12] for CF and EF. Calculations were performed in Microsoft Excel; indices were computed on per-site mean values (n = 3).

4 Results

4.1 Heavy-metal concentrations

Heavy-metal concentrations were determined in 30 replicate samples (3 per site) covering ten sequential floodplains from the source (FP1, adjacent to the coal-waste stockpile) to the most distal site (FP10, near the Qehas bridge). The full data set is presented in Table 1.

Table 1. Measured heavy-metal concentrations (mg kg^{-1}) in floodplain samples, Erzen River basin (n = 30; three replicates per site).

Sample	Coordinates	Elev. (m)	Cd	Pb	Cr	Cu
1.1s	41°13′28.03″N 19°57′40.55″E	261	91	776.4	247	422.8
1.2s			86.1	661.2	124.7	367.4
1.3s			82.5	529.7	264.9	353.6
2.1c	41°14′12.78″N 19°55′26.09″E	189	86.2	678.2	249.7	343.7
2.2c			82.6	584.7	313.6	338.5
2.3c			74.7	547	239.8	254.6
3.1	41°14′12.58″N 19°55′8.80″E	186	58.2	411.2	239.7	265.4
3.2			61.6	421.3	293.6	277.5
3.3			54.3	417.5	245.8	264.9
4.1	41°14′24.21″N 19°54′58.33″E	182	68.2	480.2	199.7	285.4
4.2			63.6	451.8	203.6	300.5
4.3			59.3	413.5	215.8	274.9
5.1	41°14′39.75″N 19°54′46.84″E	177	69.6	538.2	278.3	290.4
5.2			72.6	600.2	296.4	304.9
5.3			70.3	520.8	259.6	287.5
6.1	41°14′56.84″N 19°54′30.60″E	170	73.7	608.9	246.7	308.9
6.2			71.9	630.2	252.8	312.1
6.3			72	645.3	228.6	325.2
7.1	41°15′4.91″N 19°54′25.92″E	169	76.2	658.2	239.7	314.7

Sample	Coordinates	Elev. (m)	Cd	Pb	Cr	Cu
7.2			79.4	604.8	213.6	308.5
7.3			74.7	589.4	239.8	294.6
8.1	41°15′4.47″N 19°54′18.83″E	168	72.1	662.3	287.9	325.2
8.2			78.4	687.4	306.5	298.7
8.3			70.2	630.6	269.7	318.6
9.1	41°15′6.78″N 19°54′1.73″E	167	69.8	655.5	307.6	338.4
9.2			73.5	670.7	314.8	342.2
9.3			71.2	625.8	327.4	343
10.1	41°15′12.38″N 19°53′48.96″E	163	55	605.6	317.7	328.4
10.2			69.9	710	324.8	339.2
10.3			70.2	595	357.7	323

Coordinates and elevation reported once per site (top row).

4.2 Spatial distribution and pollution-indices assessment

Mean concentrations and pollution indices (CF, I_{geo}, EF) per floodplain are summarised in Table 2, together with the Pollution Load Index (PLI) computed for n = 4 metals. Cd, Pb and Cu display their maxima at FP1 (86.5, 655.8 and 381.3 mg kg^{-1}, respectively), decrease to minima around FP3–FP4 and rise again towards FP8–FP10, producing a broadly U-shaped longitudinal profile. The observed increase in chromium concentrations in downstream floodplain sediments may be attributed to the erosion of ultramafic lithologies within the Erzen basin and subsequent deposition of fine-grained, metal-enriched sediments under low-energy hydrodynamic conditions.

Cadmium is by far the most enriched element (CF 400–597; I_{geo} 8.06–8.64; EF 29.6–61.9), placing every site in the extreme-pollution class. Pb (CF 27.8–44.0; I_{geo} 4.21–4.87; EF 2.4–4.5) and Cu (CF 20.7–29.3; I_{geo} 3.79–4.29; EF 1.7–3.0) follow, while Cr remains in the strong-pollution class (CF 9.4–15.2; I_{geo} 2.64–3.34). PLI values, computed with n = 4 (Cd, Pb, Cr, Cu), are extreme at every site, ranging from 40.6 at FP3 to 53.4 at FP9 (mean 48.6) — more than an order of magnitude above the polluted/unpolluted threshold of 1.

Table 2. Mean heavy-metal concentrations (mg kg⁻¹), Contamination Factor (CF), Geoaccumulation Index (I_geo), Enrichment Factor (EF; reference = Cr) and Pollution Load Index (PLI; n = 4) per floodplain. CF, I_geo and PLI are referenced to the FOREGS European topsoil median.

Metal	Index	FP1	FP2	FP3	FP4	FP5	FP6	FP7	FP8	FP9	FP10
Cd	Mean	86.5	81.2	58	63.7	70.8	72.5	76.8	73.6	71.5	65
Cd	CF	596.8	559.8	400.2	439.3	488.5	500.2	529.4	507.4	493.1	448.5
Cd	I_{geo}	8.64	8.54	8.06	8.19	8.35	8.38	8.46	8.4	8.36	8.22
Cd	EF	61.87	46	33.9	46.83	38.64	45.34	50.41	38.75	34.26	29.6
Pb	Mean	655.8	603.3	416.7	448.5	553.1	628.1	617.5	660.1	650.7	636.9
Pb	CF	43.72	40.22	27.78	29.9	36.87	41.88	41.16	44.01	43.38	42.46
Pb	I_{geo}	4.87	4.74	4.21	4.32	4.62	4.8	4.78	4.87	4.85	4.82
Pb	EF	4.53	3.31	2.35	3.19	2.92	3.8	3.92	3.36	3.01	2.8
Cr	Mean	212.2	267.7	259.7	206.4	278.1	242.7	231	288	316.6	333.4
Cr	CF	9.65	12.17	11.8	9.38	12.64	11.03	10.5	13.09	14.39	15.15
Cr	I_{geo}	2.68	3.02	2.98	2.64	3.08	2.88	2.81	3.13	3.26	3.34
Cu	Mean	381.3	312.3	269.3	286.9	294.3	315.4	305.9	314.2	341.2	330.2
Cu	CF	29.33	24.02	20.71	22.07	22.64	24.26	23.53	24.17	26.25	25.4
Cu	I_{geo}	4.29	4	3.79	3.88	3.92	4.02	3.97	4.01	4.13	4.08
Cu	EF	3.04	1.97	1.75	2.35	1.79	2.2	2.24	1.85	1.82	1.68
—	PLI	52.20	50.69	40.59	40.61	47.66	48.66	48.18	51.61	53.36	52.10

5 Discussion

5.1 Transport and deposition mechanisms

The combined CF, I_{geo} and EF evidence identifies the abandoned coal-waste stockpile on the Krraba tributary as the dominant source of Cd, Pb and Cu in the ten floodplains. Coal wastes are enriched in chalcophile elements (Cd, Pb, Cu) bound to sulphide and organic phases [6–8]. The exposed stockpile undergoes oxidative weathering, erosion during high flows, and acidic runoff, releasing metal-bearing fine particles into the drainage network (Figure 4).

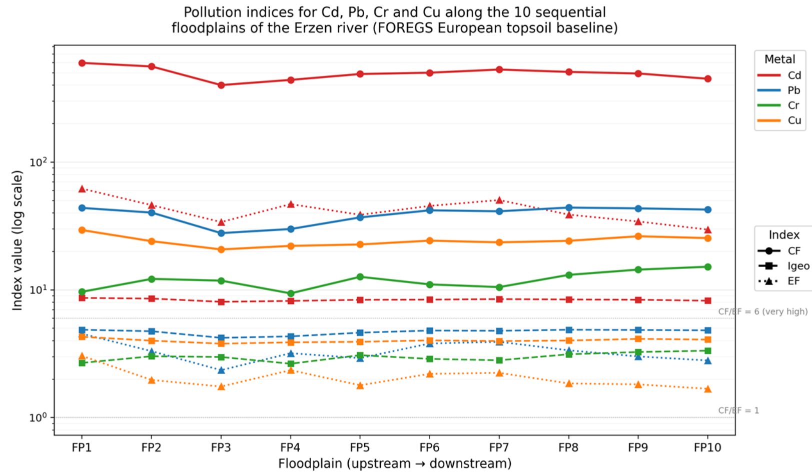

Fig. 4. Combined pollution indices (CF, I_geo, EF) for Cd, Pb, Cr and Cu along the ten sequential floodplains of the Erzen river (log Y-axis; FOREGS European topsoil baseline).

Heavy metal distribution in river sediments is controlled by anthropogenic inputs and sedimentological–hydrodynamic factors, including grain-size and organic matter content. Fine-grained sediments (silt and clay), often associated with organic-rich fractions, show higher metal adsorption capacity than coarser materials.

Hydrodynamic conditions (discharge, flow velocity, channel morphology) control sediment transport, sorting, and deposition, with preferential metal accumulation in low-energy floodplain environments dominated by fine sediments.

The U-shaped longitudinal profile of Cd, Pb and Cu reflects a two-stage transport process typical of mid-sized Mediterranean basins [3]. It begins with deposition of coarse particles near the source (FP1–FP2) and continues with accumulation of metal-rich fines in slack-water zones (FP3–FP4). During successive high-flow events (FP5–FP10), these sediments are progressively remobilized, mixing with lithogenic materials that carry a higher chromium (Cr) signature transported from upstream [5]. Meanwhile, elements such as Cd and Pb, due to their high affinity for fine organic particulates and Fe/Mn oxyhydroxides, remain strongly bound to the solid phase, exhibiting minimal attenuation along the ~4 km monitoring reach [1, 3].

5.2 From a localized source to distributed contamination and management implications

PLI values remain extreme at all floodplains (40.6–53.4 for n = 4 metals), showing how a point source (~219,252 m³ of coal waste) can generate severe contamination along a ~4 km river corridor compared to the European FOREGS baseline [10]. The affected floodplain area (≥7.3 ha) acts as a long-term secondary reservoir of Cd, Pb and Cu, with potential remobilisation during future floods even after source removal.

Because Cd and Pb enrichment persists downstream to FP10, ecological receptors and local communities may remain exposed, indicating potential ecological risk not directly quantified here [2, 4]. Local observations from Krraba and Ibë indicate ecological changes, including reduced riparian vegetation (e.g., *Rubus* spp., *Salvia officinalis*, *Thymus vulgaris*, *Origanum vulgare*) and a reported decline of *Barbus albanicus* over a ~3 km reach. Although not systematic, these observations provide qualitative evidence of potential ecological stress.

The absence of systematic floodplain monitoring and rehabilitation in Albania contributes to persistence of these secondary sources [4,5]. Key recommendations include: (i) inclusion of floodplain soils in monitoring alongside riverbed sediments; (ii) integrated use of CF, I_{geo}, PLI and EF against European baselines; and (iii) remediation of abandoned coal-waste stockpiles (capping, re-vegetation or removal) with land-use restrictions in adjacent floodplains. Implementation within the EU Water Framework Directive framework [9] would reduce long-term environmental risks.

6 Conclusion

A single localised point source — an abandoned coal-waste stockpile on the Krraba tributary — has generated a sequence of severely contaminated floodplains along a ~4 km reach of the Erzen river. Against the harmonised FOREGS European topsoil baseline [10], Cd and Pb reach extreme levels at all ten sites (CF up to 596.8; I_geo up to 8.64), Cu remains persistently elevated (CF 20.7–29.3; EF 1.7–3.0), and Cr behaves as a lithogenic element with downstream enrichment from ultramafic detrital inputs. PLI values remain above 40 at every site, confirming that a small point source has generated a diffuse, cumulative secondary-pollution footprint over several hectares of alluvial soil.

Floodplain soils must be integrated into systematic monitoring and remediation programmes in Albania, as they currently act as hidden long-term reservoirs of heavy metals. Floodplain-focused geochemical surveys, complemented by multiple pollution indices and referenced to a harmonised European baseline, provide a robust decision-support tool for sustainable river-basin management in mining- and coal-affected catchments.

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