

Sewage Sludge and Sediment Analyses as Indicators of Regional Heavy-Metal Pollution: A Case Study in El-Harrach, Algeria

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Abstract

This study examines pollution in the El-Harrach region in northern Algeria by analyzing concentrations of heavy metals and trace elements in sewage sludge and sediments. Three locations were sampled: upstream, within the Baraki wastewater treatment plant (WWTP), and downstream. To identify the major mineralogical components and pollutant composition, chemical analyses were performed using advanced methods, including X-ray diffraction (XRD), X-ray fluorescence (XRF), Inductively Coupled Plasma Mass Spectrometry (ICP-MS), Atomic Emission Spectroscopy (ICP-AES), and total organic carbon (TOC) analysis. Sediments were heavily enriched in heavy metals, particularly at the WWTP. This location yielded concentrations of 2200 ppm copper (Cu), 1160 ppm chromium (Cr), 1590 ppm zinc (Zn), and 320 ppm nickel (Ni). In the downstream areas, lead (Pb) and arsenic (As) were present at 330 ppm and 60 ppm, respectively. The upstream samples showed lower levels, with 410 ppm Cu, 680 ppm Cr, and 100 ppm Ni. Pollution level indicators, including the contamination factor (CF) and enrichment factor (EF), indicated extreme signs of pollution, primarily due to Cu, Zn, Cr, and As, resulting from the influence of industrial effluent and WWTPs. These findings indicate the need for continued environmental monitoring and implementation of effective remediation programs to mitigate these environmental and public health risks in the region.

Keywords:

Heavy metals, Contamination, Oued El-Harrach, Industrial discharges, Environmental pollution.

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1. Introduction

Heavy metals are toxic metallic elements with a density above 5 g/cm³ (Fergusson and Prucha, 1990; Zhao et al., 1998; Chen et al., 2015) and are generally represented by having an atomic number above 20 in the periodic table. These elements, such as copper (Cu), lead (Pb), chromium (Cr), zinc (Zn), nickel (Ni), and arsenic (As), exist in trace quantities under natural environmental conditions (Yildiz and Özkul,

2024). However, they are irreversible environmental pollutants caused by natural processes as well as a host of human activities. Uncontrolled heavy metal leakage into the land and rivers is a significant environmental problem. They are hazardous and cannot be broken down into non-toxic forms, causing a global health issue (Al-Khafaji and Jaafar, 2023).

Heavy metals are also found in association with fine-grained sediments and sludge, especially in organic-rich substrates (Robbe, 1981; Marchand et al., 2006; Banfalvi, 2011; Blais et al., 2005;

Atroune, 2012; Power and Chapman, 2018). Sediments serve as a container for metals present in the environment (Ouattara et al., 2021; Takahashi et al., 2023). The study of sediments thus makes it possible to understand the environmental changes that have taken place in river basins and offer valuable information on the accumulation of heavy metals (Leppänen and Kukkonen, 2004), thus reflecting the natural (geogenic) and human consequences (Konan et al., 2021), because soil pollution by trace metal elements has become an environmental problem of concern globally, raising concerns about its impacts on human health, biodiversity and the sustainability of terrestrial ecosystems (Masindi and Muedi, 2018). In the south of the Mediterranean, in France, the Marseille wastewater treatment plant is one of the largest in Europe (Ricou, 2023). Barcelona has implemented advanced wastewater treatment technologies in Spain, integrating biological and chemical processes to recycle water and reduce ecological impact (Silva, 2021). Italy, on the other hand, allows for efficient reuse of treated water for agricultural irrigation (Garrido-Cardenas et al., 2019). These countries are showing sustainable and innovative solutions to improve water quality and protect marine ecosystems.

In Algeria, the population and industry are concentrated in the north of Algeria and overlook the Mediterranean Sea, which has become a direct or indirect discharge (exoreic basin) for the wadis loaded with wastewater and untreated industrial liquid discharges. These wadis flow towards the north because of the slope of the Atlas chain, and they bring with them the ETMs that are often present there. Studies have already been conducted on sludge from wastewater treatment plants and ETMs in inland areas (Boudraa, 2020). On the other hand, in coastal and overpopulated areas such as the Wilaya of Algiers, the first study established that, given the volume of different wastes discharged without treatment, the Oued El Harrach, an open sewer, evacuates various pollutants. In addition, the Odors it emits disturb the living environment of the residents of the city of El Harrach (Atroune et al., 2012), caused by several factors such as high population density and intensive industrial activity, which increase the emissions of these elements into the atmosphere and surrounding waters (Mahia, 2015).

Although the Baraki wastewater treatment plant (WWTP) receives a large share of effluent from the urban and industrial networks of the wilaya of Algiers (Ferradj and Idouhar, 2016), it still does not remove all contaminants, including trace elements, and the effluent still does not comply with discharge standards for the natural environment (Louati, 2015). But it does not remove all types of contaminants, including trace elements. The objective of this study is to assess the status, the risk of pollution, and the potential sources of pollution in elements Fe, Pb, Cr, Ni, Cu, Zn, As, and Ga in the El-Harrach region of northern Algeria through the analysis of sediments upstream and downstream and the sewage sludge derived from the wastewater treatment plant.

2. Description of the Study Area

The watershed of Oued El-Harrach maritime is located in the Mitidja Oriental plain, with an area of 835.8 km², a perimeter of 142 km, and an altitude between 20 and 1629 m (Ammari, 2012; Bouchelouche et al., 2019). This watershed occupies nineteen communes in the central area of the Wilaya of Algiers, with a population of 1,200,000 inhabitants and a wastewater discharge of 107,569.67 m³. The industrialization in this watershed is concentrated along the Oued El-Harrach and consists of three major industrial zones: the Oued Smar zone, the El Harrach zone, and the Ford zone of Constantine (Haouchine, 2010). The area studied is part of the sub-watershed from Beni Talha, south of Algiers, to Al-Sablât in the north via the Baraki wastewater treatment plant (Figure 1).

At the climatic level, this sloping basin is part of the Mediterranean region, with hot, humid winters and cool, dry summers. The average annual temperature is about 18 °C, with a maximum temperature of 30 °C during the summer, while the average annual precipitation is between 600 and 800 mm (Hamitouche et al., 2024). The average annual evaporation is estimated at 865 mm (Hadjji and Belhocine, 2021).

The region of El Harrach and Algiers has a rich and varied geology, mainly shaped by tectonic and sedimentary events. El Harrach, located east of Algiers, is in the Italian zone of Algeria and is known for its complex structures resulting from the convergence between the African and Eurasian plates (Ficheur, 1901). This region is characterized by geological formations dating from the Mesozoic

and Cenozoic periods, including a series of flysch, marl, and limestone formations. The watershed of the Oued El-Harrach dominates the Mitidja plain. This area is largely made up of red sandy silt formations of superficial Plio-Quaternary age,

about ten meters thick, resting on Tertiary and Cretaceous carbonate formations. To the west, the watershed is bounded by Cretaceous metamorphic and flysch basement (Zouakh and Bouhadad, 2013).

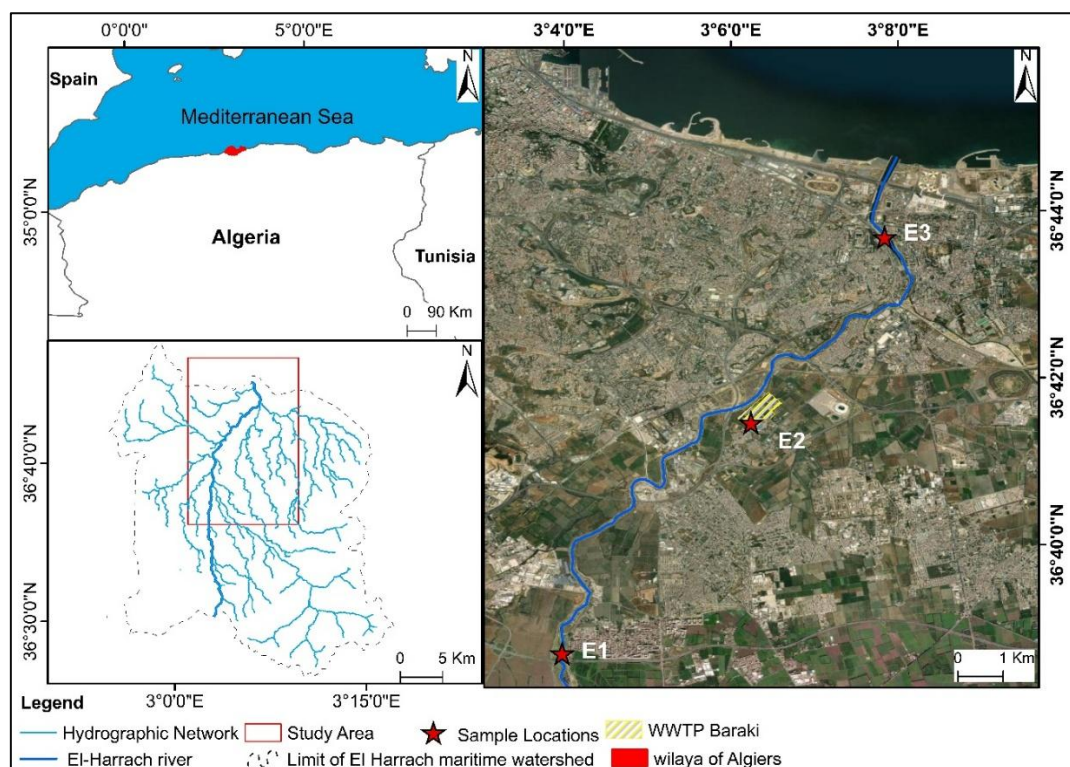


Fig. 1: Location and geographical location of the study area. (E1: Upstream, E2: Center, E3: Downstream.)

3. Materials and Methods

A. Sampling

After conducting exploration in the southern marine catchment area of Wadi El-Harrach and to accurately represent the study area, samples were taken from three sites in Wadi El-Harrach at a distance of 14 km and a depth of 20 cm from the fine sediments (silt) at the upstream E1 and downstream E3; other samples were taken in the middle of the E2 wastewater treatment plant (liquid sludge of Oued El Harrach) (Baraki wastewater treatment plant), each weighing 1.5 kg. The locations of these samples were determined by the Global Positioning System (GPS) and reported in the analytical maps. The liquid sludge was collected using a hand shovel, packed into sterile plastic boxes, numbered, transported in a cooler, and then stored in a laboratory refrigerator.

B. Geochemical analysis

Analysis by ICP-AES and ICP-MS

Three (3) representative samples were sent for geochemical analysis (major elements, trace elements, and rare earths) to the ALS Minerals laboratory in Seville, Spain. The method used for major elements, trace elements, and rare earth elements is an ultra-trace level method using Inductively Coupled Plasma Spectroscopy—Atomic Emission (ICP-AES) called "ME-MS61r". A prepared sample (0.25 g) is digested with perchloric, nitric, hydrofluoric, and hydrochloric acids. The residue is diluted with hydrochloric acid and analyzed by inductively coupled plasma atomic emission spectrometry.

X-ray diffraction (XRD) analysis

A 1 g sample of raw material was crushed to a particle size of 150 micrometers and then analyzed using an X-ray diffractometer (XRD)

"Olympus BTX-716 Benchtop XRD" equipped with a Cu-K α anticathode ($\lambda = 1.540598\text{\AA}$).

The analysis was conducted at the Sahara Geology Laboratory of the University of Ouargla. The X-ray spectra of all samples were recorded in the range of $2\theta = 5^\circ$ to 55° , with a step size of 5° at a scanning speed of $2^\circ/\text{min}$. The obtained data were compared with the model registered in the PDF2 database using PANalytical X'Pert HighScore Plus software.

X-ray fluorescence (XRF) analysis

The samples underwent pre-treatment at the chemistry laboratory of the University of Constantine 1. The liquid sludge was air-dried for 48 hours at room temperature. After complete drying, the samples were finely ground using a Retsch PM 100 mill for 45 minutes at 320 rpm, resulting in a powder with a particle size less than 100 nm. Subsequently, 10 grams of the powdered sample from each set were compressed into a 30 mm diameter pellet using a Retsch PP 25 manual pelletizer at a pressure of 10 tons. This was done to facilitate X-ray fluorescence (XRF) analysis using a Panalytical Epsilon 3 spectrophotometer available in the Pollution and Water Treatment Laboratory, Department of Chemistry, University of Constantine 1.

For accurate analysis, XRF data of all samples were recorded under different excitation conditions of DDP and current: 5.00 kV and 1000 μA , 30.00 kV and 300 μA , 12.00 kV and 519 μA , 20.00 kV and 450 μA , respectively. X-ray fluorescence (XRF) is particularly effective for detecting heavy metals in solid matrices, while

inductively coupled plasma atomic emission spectroscopy (ICP-AES) allows for a broader analysis of elements, including transition metals, with increased sensitivity in complex sample matrices

Total organic carbon (TOC) analysis

To determine the total organic carbon concentration of the soluble matter in the samples, 10 g of crushed slurry is mixed with 100 ml of distilled water. After stirring the mixture for an hour, it is filtered through filter paper. Then, 10 ml of the solution from the last step is mixed with 90 ml of distilled water. The new solution is then tested with an Analytik Jena Multi N/C 3100 apparatus available at the Polytechnic Laboratory of Constantine University 3.

Assessment and monitoring of contamination

To assess the level of trace-element contamination in sediments (pollution indices), it is necessary to determine parameters such as the enrichment factor, contamination factor, geo-accumulation index, degree of contamination, and sediment pollution index. This information is essential for implementing appropriate measures to mitigate the negative impacts of pollution and preserve ecosystem biodiversity. The calculations were performed according to the protocols of Buzea et al. (2007), Kowalska et al. (2018), Al-Janabi et al. (2019), and Okerefor et al. (2020) to assess the environmental impact of pollution (Table 1). These parameters allow quantification of heavy-metal and other trace-element concentrations in sediments and comparison with reference values from Wedepohl (1995).

Table 1: Pollution indices and covariates utilized in this study.

Indices	Definition	Formulas	Classification
Contamination Factor (CF) (Hakanson, 1980)	The Contamination Factor (CF): considers the relative level of pollution in the soil.	$CF = C_N / C_B$ C_N : concentration of the element in the sample C_B : concentration of the background element	$FC < 1$: Low contamination $1 \leq FC < 3$ Moderate contamination $3 \leq FC < 6$ Considerable contaminations $FC \geq 6$: Very high contamination
Enrichment Factor (EF) (Zoller, 1974) Classification Birch and Olmos (2008)	The enrichment factor (EF) is defined as a quantitative measurement used to assess the increase in the concentration of an element in the soil.	$FE = (C_N / C_{Fe})_E / (C_N / C_{Fe})_B$ $C_N / C_{Fe})_E$: the ratio of the concentration of a C_N element and the concentration of iron C_{Fe} in the sediment $(C_N / C_{Fe})_B$: the ratio of the concentration of a C_N element and the concentration of C_{Fe} iron in the background.	$EF \leq 1$ No Enrichment $1 < EF \leq 3$ Minor Enrichment $3 < EF \leq 5$ Moderate Enrichment $5 < EF \leq 10$ Moderately severe enrichment $10 < EF \leq 25$ Severe Enrichment $25 < EF \leq 50$ Very severe enrichment $50 \leq EF$ Extremely high enrichment $0 < I_{Geo} < 1$ Not contaminated
$I_{geo} = \log_2(C_N / 1.5 \times C_B)$			

Geo-accumulation Index (I Geo) (Müller, 1969) Classification (Förstner, 1969)	Geo accumulation index (Igeo): is the presence of metals in the soil compared to the natural bottom level as a reference.	C_N : concentration of the element in the sample C_B : background element concentration 1.5: background exaggeration factor	$1 < I_{Geo} < 2$: Not contaminated too $2 < I_{Geo} < 3$ Moderately contaminated $3 < I_{Geo} < 4$ Moderately to severely contaminated $4 < I_{Geo} < 5$ Severely to extremely contaminated $5 < I_{Geo}$ Extremely contaminated.
Degree of Contamination (DC) (Hakanson, 1980)	Degree of contamination (DC): is defined as the level relative to the level of pollution by trace elements in the soil.	$DC = \sum FC$	$CD < 8$ Low $8 \leq CD < 16$ moderate $16 \leq CD < 32$ considerable $32 \leq CD$ Very high
Sediment Pollution Index (SPI) (Singh et al,2002).	Sediment pollution index (SPI): described as a parameter used to assess the degree of contamination of sediments by toxic substances,	$IPS = \sum (EF_m \times W_m) / \sum W_m$ W_m : toxicity weight or weighting factor for metal m. A weight of 1: Cr and Zn, 2: Cu and Ni, 5: Pb. EF_m : enrichment factor for metal m	$0 \leq IPS < 2$ Sound sediment. $2 \leq SPI < 5$ Slightly polluted sediment. $5 \leq SPI < 10$ Moderately polluted sediment. $10 \leq SPI < 20$ Heavily polluted sediment. $SPI \geq 20$ Hazardous sediments.

4. Results and discussion

A. Geochemical composition of the sludge

A. Analysis of the X-ray diffraction spectra (Figure 2) of sludge from three study sites (E1, E2, and E3) indicates a mineralogical composition dominated by quartz (SiO_2), alumina (Al_2O_3), and calcite ($CaCO_3$), as well as several clay and metal-bearing phases. These minerals were identified based on their characteristic peaks using the corresponding ASTM PDF sheets (00-001-0649, 01-072-1937, and 00-004-0878). Quartz, present in all samples, shows peaks at 20.72° , 26.65° , and 49.87° for the E1 sample; at 20.84° , 26.58° , and 50.01° for E2; and 20.76° , 26.49° , and 49.85° for E3, indicating a significant detrital input linked to the erosion of surface formations. The presence of calcite ($CaCO_3$) is identified by its peaks at 29.44° , 35.97° , 47.33° , and 48.49° for E1, at 29.33° , 35.93° , 47.44° , and 48.43° for E2, and at 29.22° , 35.97° , 47.27° and 48.23° for E3, resulting from the leaching of Cretaceous carbonate rocks. The alumina oxide (Al_2O_3) peaks at 13.79° , 32.25° and 34.88° for E1; at 13.82° , 32.30° , and 34.83° for E2; and at 13.74° , 32.51° , and 34.79° for E3. These are attributed to the geological alteration of aluminium-rich minerals and the dissolution of rocks containing aluminous phases. The presence of albite and clay minerals such as kaolinite, illite, and vermiculite reflects advanced alteration processes and marked pedogenetic dynamics (Kabata-Pendias, 2010).

Beyond this classic mineralogical composition, metal-bearing phases such as chalcopyrite ($CuFeS_2$) and sinnerite ($Cu_6As_4S_9$) were also identified. As part of the study of anthropogenic inputs, particularly industrial ones, the analysis of arsenosulfanite ($Cu_3(As, V)S_4$), renierite, helvite, and litharge (PbO) provides a clear indicator of these inputs. These phases, which are not common in an ordinary sedimentary context, are often associated with trace metals such as Cu, Zn, Ni, Cr, Mn, and As, which can be used to trace their origins and propose the technique of their treatment (Fakhru et al., 2009; Boudraa et al., 2017). Their presence is typically linked to metallurgical or chemical discharges (Sundaray et al., 2010). Clay minerals and oxides (Fe, Mn, Al) play an essential role as geochemical traps. They act by adsorption or coprecipitation. This contributes to the partial immobilization of these elements in the sedimentary matrix (Förstner and Wittmann, 1981).

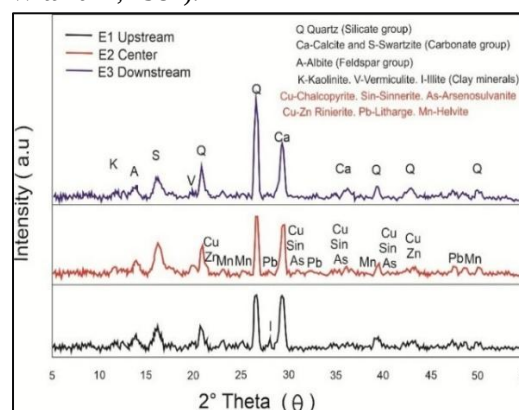


Fig. 2 X-ray diffractometry spectra of the sludge analyzed.

Semi-quantitative analysis of samples E1, E2 and E3, carried out by inductively coupled plasma mass spectrometry (ICP-MS) and inductively coupled plasma spectroscopy -Atomic Emission (ICP-AES), reveals a mineral composition dominated by silicon, aluminium, calcium and iron oxides. The respective contents vary between 18.8–42.5% for SiO₂, 6.03–11.85% for Al₂O₃, 11–16% for CaO, and 3.21–5.79% for Fe₂O₃ (Table 2). The presence of SiO₂ and Al₂O₃ shows a composition of aluminosilicate resulting from the alteration and transport of detrital components of recent sandy-silty formations that cover the watershed studied. The dominance of these minerals, SiO₂, Al₂O₃, and Fe₂O₃, is reported by the work of Foroughi et al. (2018) and Kouassi et al. (2022).

Compared to the results of the sludge study in Zambia by Phiri et al. (2021), who reported quartz contents ranging from 10.5% to 19.1% in sludge effluents and solid deposits, the higher SiO₂ concentrations observed in this study (18.8% to 42.5%) indicate a significantly greater siliceous enrichment in the Oued El Harrach samples. This elevated quartz content suggests a more pronounced presence of siliceous minerals, likely reflecting differences in geological sources or environmental conditions between the studied locations. These findings underscore the importance of local geochemical and sedimentological factors in determining mineral composition in sediment samples.

Also, the high levels of total organic carbon (TOC) show that a lot of organic matter has built up from both natural sources (like algae growth and plant debris) and human-made sources, especially because treated or poorly treated domestic and industrial waste is still being released. The dual origin of organic matter is regularly seen in river systems that are influenced by urban and industrial activities (Vuillemin and Le Réveillé, 2001). The organic matter present can play a decisive role in the mobility or retention of trace metals, particularly through complexation, adsorption, or chelate formation, thus influencing the geochemical dynamics of pollutants in sediments.

Table 2: Chemical composition of sludge (% by weight).

Elements	E1	E2	E3
SiO ₂	34	18.8	42.5
Al ₂ O ₃	11.85	6.03	8.48
Fe ₂ O ₃	4.87	3.21	5.79

CaO	16	11.05	16.6
MgO	1.15	1.11	1.06
Na ₂ O	0.69	0.38	0.65
K ₂ O	1.31	0.88	1.16
Cr ₂ O ₃	0.018	0.022	0.028
TiO ₂	0.59	0.38	0.4
MnO	0.04	0.03	0.05
P ₂ O ₅	0.63	0.2	0.23
SrO	0.04	0.03	0.16
BaO	0.04	0.07	3.04
LOI	26.8	53.8	18.4
total	98.028	95.992	98.548
TOC	8.42	9.9	10.62

B. The concentration of target elements in the sludge

This analysis is crucial to understand their composition and assess their environmental impact. The results of the (ICP-MS) and (ICP-AES) analyses (Table 3) of the sludge show remarkably high concentrations of industrial elements (trace elements and primary elements), such as aluminium (Al), calcium (Ca), phosphorus (P), potassium (K), sodium (Na), magnesium (Mg), strontium (Sr), and chromium (Cr), in the three types of sludge, showing notably high concentrations of calcium and aluminium. Calcium levels reached 381250 ppm and aluminium levels 106050 ppm, far exceeding the background reference values of 29450 ppm for Ca and 77440 ppm for Al, indicating strong enrichment. Comparable studies on sewage sludge, such as Suanon (2017) in China, reported maximum values of 44800 ppm for Al and 20000 ppm for Ca, confirming the consistent accumulation of these elements in sludge over time. These elevated concentrations are likely due to mineral contributions from industrial effluents and urban discharges (Westerhoff et al., 2015; Yaseen and Scholz, 2018). Additionally, the significant concentration of vanadium (V) in the upstream sample (123 ppm in E1) may reflect specific industrial inputs or geogenic sources mobilized by anthropogenic activity.

The precious elements detected in the three samples analyzed (Table 3) generally exhibited negligible concentrations in the upstream and downstream. However, the distribution of the element ruthenium (Ru) exhibited a clear pattern of enrichment, where the downstream area showed the maximum value (1.66 ppm in E3), much higher than the background value of the Upper Continental Crust of approximately 1×10^{-4} ppm and also higher than the highest value of 0.110 ppm thus far recorded in sewage sludge by Suanon

(2017). This clear indication of enrichment points to spatial variation in the concentration of precious elements that cannot be explained by anything other than industrial effluents, as well as urban technological activity, producing the downstream load.

The rare earth elements (REE) identified in the sludge samples exhibit concentrations generally similar to or slightly lower than those of the background (Wedepohl, 1995), with certain enrichments around stations. Concentrations in the upstream sample (E1) vary between 0.14 mg/kg for lutetium (Lu) and 47.3 mg/kg for cerium (Ce), while the downstream values (E3) reach 36.9 mg/kg for Ce and 29.1 mg/kg for yttrium (Y). Relative to background, elements like lanthanum (La), yttrium (Y), and gadolinium (Gd) show an enrichment factor of more than 1, reflecting an accumulation in excess of natural geochemical background levels. Conversely, heavy rare earth elements like the elements ytterbium (Yb), thulium (Tm), and lutetium (Lu) exhibit minimal depression. The distribution pattern is indicative of both the natural lithogenic inputs as well as an anthropogenic component. The enrichment in La, Y, and Gd, especially at the downstream station, points towards the introduction of REE-bearing particles resulting from industrial and urban activity, among them the activity of metallurgy, battery manufacturing, as well as electronic waste. These results agree with the findings presented by Pourret and Hursthouse (2019), who explained the occurrence of REEs in watery environments as resulting from the remobilization of small fractions of particles, leaching of enriched soils, as well as industrial effluents.

Table 3: Concentrations (in ppm) of elements detected in the sludge studied.

Elements	E1	E2	E3
Industrial elements			
AL	106050	70180	84670
Ba	352	605	>10000
Ca	349870	381250	347370

P	4170	24420	950
K	34210	33110	26750
Na	5480	2980	4560
Mg	10520	13600	9450
W	1.8	1	1.4
Sr	421	338	1445
Sn	5.2	8.6	14.8
Cr	128	152	204
Ga	18.6	8.9	12.6
V	123	57	82
Rb	66	36.7	56.9
Nb	12.25	6.37	7.6
Hf	2.61	2.57	2.78
Precious elements			
Ru	1.03	0.59	1.66
Rare earth elements			
Ce	47.3	30.1	36.9
Nd	19.6	14.2	18.5
La	37.2	18	35.5
Y	19.3	27.7	29.1
Sm	3.92	2.96	3.71
Yb	1.56	0.87	1.26
Dy	2.78	2.01	2.93
Er	1.92	1.02	1.66
Ho	0.53	0.39	0.6
Tb	0.5	0.35	0.44
Tm	0.22	0.13	0.2
Lu	0.14	0.17	0.16
Pr	5.62	3.37	4.26
Gd	3.59	2.36	3.87

The relative amounts of different elements encountered in the analyzed samples (Fig. 3) show that the upstream sample (E1) is notably enriched with the elements Al, Ba, Ca, and K. In contrast, the downstream sample (E3) has the highest accumulation of elements, which suggests that elements from upstream springs and sludge in the center of the treatment plant accumulate downstream; this confirms that some elements are natural. They are the result of the alteration of the different rocks (leaching) of the mountains, and others are anthropogenic, agricultural, and industrial (Pourret and Hursthouse, 2019; Latosińska et al., 2021; Witkowska et al., 2021; Abbasi et al., 2024). Chemical and nutrient residues in treated wastewater show that the treatment process does not entirely remove these elements, which explains their presence in WWTP sludge (Suanon et al., 2017; Leppänen et al., 2024).

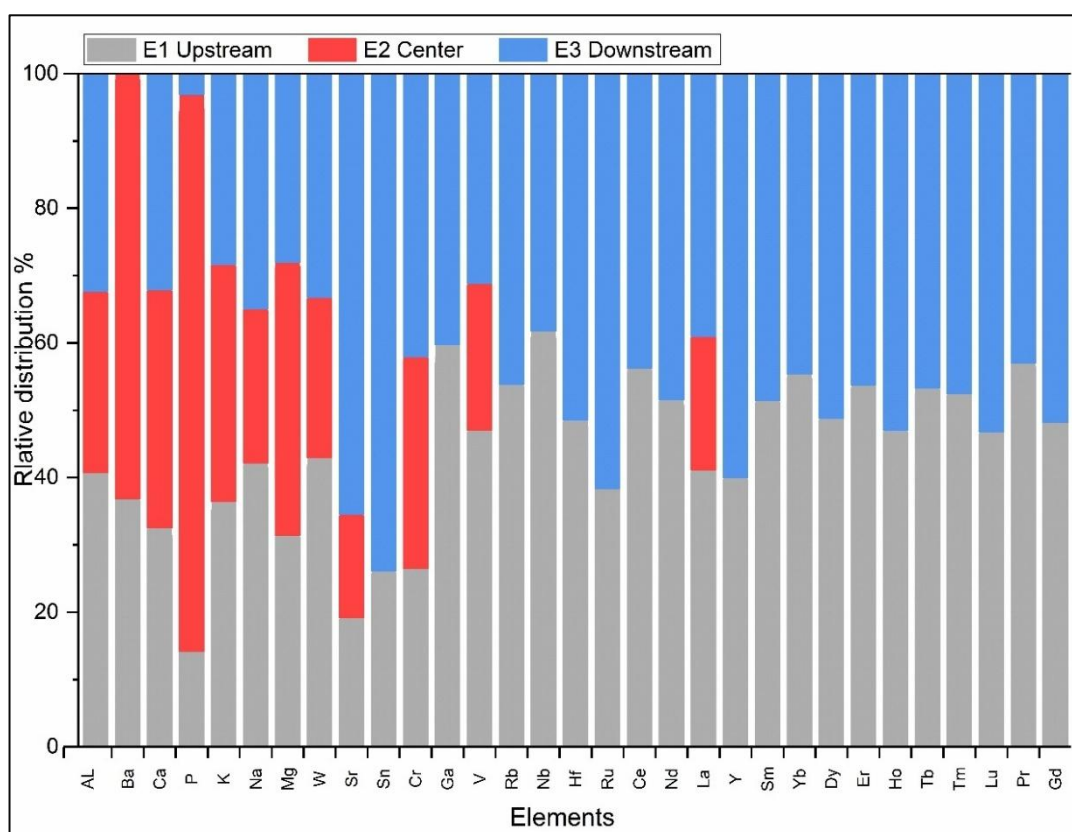


Fig. 3: Spatial variations in the relative distribution percentages of the target elements.

C. Pollution indices

Contamination factor (CF)

The analyzed sediments have a considerable number of metals. So, it is necessary to classify and quantify these metals to have an idea of their contamination rates and to investigate their origins. The results of the analyses (Figure 4a) show strong contamination ranging from 12 to 28 in all sediments by the metals Zn, Cr, Cu, and As; an extreme value of 153 for Cu in the WWTP sludge; and Ni is also present in the WWTP sediments with a value of 17.2. At the same time, Pb reaches a value of 19.41 downstream of Oued El Harrach. These values exceed the reference thresholds established by Hakanson (1980) ($CF \geq 6$). The high values of CF are due to two main reasons: the first is the treatment press machine at the stations that concentrates metals in the sludge, and the second reason is due to the discharges of industrial activity near Oued El Harrach (De Fátima Braga Barbosa et al., 2019; Murillo et al., 2021), which gives rise to different types of industry such as battery production. Considerable contamination ($3 \leq FC < 6$) has been observed for the elements Ti and Pb in samples taken upstream and at the WWTP, with

values between 3.5 and 4.5. For Ni, values of 5.38 and 4.30 were recorded, respectively, in the upstream and downstream sediments. Meanwhile, Ga has a value of 3.57 in the upstream sediments, and Mn reached 3.52 in the WWTP sediments. Moderate contamination is also observed for Mn in upstream and downstream sediments, with values of 2.37 and 2.08, respectively. For Ga, values of 2.14 and 1.42 were recorded, respectively, in the WWTP and downstream sediments, while Ti has a value of 2.93 in the downstream sediments. In contrast, Fe has low contamination and is considered a mineral is observed with a value of 2.46. Moderate contamination ($1 < I_{geo} < 2$) of upstream Ni is observed, with a value of 1.07. In contrast, the geo-accumulation indices of Fe, Ti, Mn, and Ga in samples upstream and in the center of the treatment plant show no to moderately contaminated ($0 < I_{geo} < 1$). This suggests that the sediment is not contaminated with the elements mentioned.

Enrichment factor (EF)

The calculated enrichment factors reveal extremely severe enrichment ($EF \geq 50$) in metals according to the classification of Birch and Olmos

(2008). Cu values are 5.15 upstream and 283.94 at the WWTP, while Zn and Cr reach 56.43 and 61.17, respectively, in the WWTP sediments. Downstream, As has a value of 67.08 (Figure 4b). A *very severe enrichment* ($25 < FE \leq 50$) has been observed in Zn and Cr in the sediments upstream and downstream, with values between 27.47 and 48.75. Cu and Pb show values of 39.09 and 43.4, respectively, in downstream sediments, while As shows values of 40.56 and 46.14 in upstream and WWTP samples. Ni has a value of 31.75 in the sludge from the water treatment plant. *Severe enrichment* ($10 < FE \leq 25$) is observed for Ni downstream, with a value of 10.9. *Moderately severe enrichment* ($5 < FE \leq 10$) was observed for Ti in all samples, as well as for upstream Pb and at the WWTP, with values of 7.15 and 6.51. Ga has a value of 7.24 upstream, while Mn is observed at the WWTP, and Ni is recorded downstream of the El Harrach wadi. *Moderate enrichment* ($3 < FE \leq 5$) for upstream and downstream Mn, as well as for Ga, is poorly enriched compared to the geochemical background values generated by Wedepohl (1995) in the WWTP and downstream.

Geo-accumulation index (I Geo)

Calculations of the geo-accumulation index for each metal at the different sampling points according to the classification of Müller (1969). *Extreme contamination* ($5 < I_{Geo}$) was observed for Cu upstream and at the wastewater treatment plant (WWTP) with values of 5.75 and 30.87, respectively, as well as for As in the WWTP and

downstream with values of 5.07 and 6.02, and for Zn and Cr in the WWTP sludge (Figure 4c). Contamination ranging from *severe to extreme* ($4 < I_{Geo} < 5$) for Zn upstream and downstream, with values of 4.82 and 4.24, and for As upstream with a value of 5.01. *Severe contamination* ($3 < I_{Geo} < 4$) of upstream Cr with a value of 3.89, Ni in WWTP mud with a value of 3.45, and downstream Cu and Pb with values of 3.50 and 3.89, respectively. *Moderate to severe contamination* ($2 < I_{Geo} < 3$) for downstream Cr of anthropogenic origin (Hernandez et al., 2003; Fatmi et al., 2023).

Degree of contamination (DC) and sediment pollution index (SPI)

The degree of contamination and the sediment pollution index provide information on the contamination of the sediments analyzed. Based on the calculation of the degree of contamination and the sediment pollution index (Anbalagan, 2023), sediments are classified according to their level of metal contamination. According to the classification of Hakanson (1980), the results of the sediment contamination degree are very high $32 \leq DC$ with values of 111.53 and 111.85 upstream and at the WWTP (Figure 4d), and an extreme value downstream of 273.59. The IPS sediment pollution index, according to Singh et al. (2002), classifies all sediments in a $20 \leq IPS$ hazardous zone with values of 23.82, 71.05, and 35.39. Upstream of the WWTP and downstream of Wadi El Harrach.

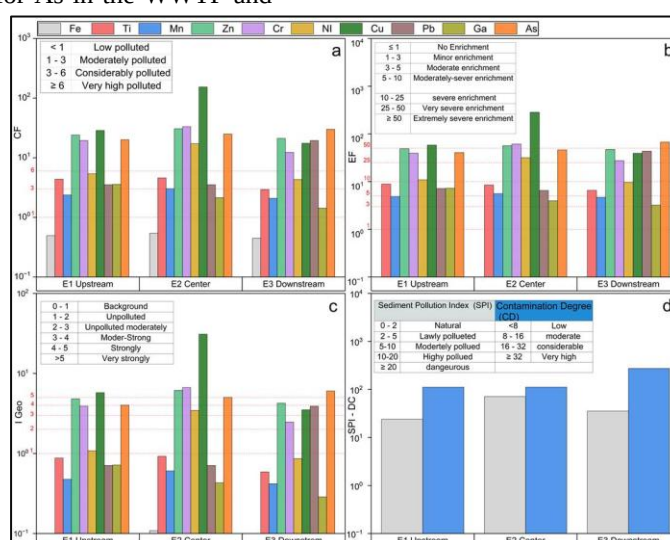


Fig. 4: a: Variations in the contamination factor in sediments; 4b: Enrichment factor (EF) values calculated for sediments according to the classification of Birch and Olmos (2008); 4c: Calculated geo-accumulation index values for trace elements in sediments; 4d: Calculated contamination degree and Sediment Pollution Index values for sediments.

5. Conclusion

This study provides compelling evidence of significant contamination in the El-Harrach region of northern Algeria, where industrial activities and high-density urban settlements contribute to substantial pollution loads. Geochemical and mineralogical analyses of sediments reveal the presence of essential minerals such as quartz, albite, calcite, and clay minerals (vermiculite, kaolinite, and illite), alongside secondary mineral phases incorporating heavy metals, including chalcophyrite, sinnerite, arsenosulvanite, renierite, helvite, and litharge. The presence of elevated levels of industrial provenance elements (Al, Ca, P, K, Na, Mg, Sr, Cr) and rare earth elements in WWTP sludge and river sediments is above the natural geochemical background levels and therefore indicates a strong anthropogenic impact. Various pollution indices (CF, I Geo, EF, CD, SPI) also indicate strong enrichment and severe contamination by Cu, Zn, Cr, and As, indicating that these sediments belong to the hazardous group. The inefficiency of the WWTP in managing heavy metal loads, particularly during periods of high rainfall and flooding, presents an urgent need for enhanced mitigation measures. Research points to the need for introducing advanced technologies in wastewater treatment processes. Stricter controls on industrial emissions and regular checks on sediment quality could make a real difference too. Collaboration among the Ministry of Environment, industry players, and research institutions seems crucial here. It would allow for ongoing evaluations and better handling of heavy metal pollution overall. Such joint actions might gradually reduce the pollution now affecting the Oued El Harrach basin. In the end, they could safeguard water quality and the broader environmental health for years to come

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7. Conflict Of Interest

The author, BOUSSELIU Amina, declares that there are no conflicts of interest regarding the publication of this paper titled "Sewage Sludge and Sediment Analyses as Indicators of Regional Heavy-Metal Pollution: A Case Study in El-

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This statement ensures transparency regarding any potential conflicts and aligns with typical journal requirements for conflict-of-interest disclosures.

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9. Author Contributions

(1) Amina. Bousseliou, as the first corresponding author, conducted comprehensive geological fieldwork and was responsible for collecting sediment samples from the El-Harrach River, as well as sludge samples from the wastewater treatment plant. She also performed extensive laboratory analyses. She processed and interpreted the analytical data and made a substantial contribution to the scientific interpretation of the results. She prepared the initial draft of the manuscript and incorporated subsequent revisions based on valuable feedback from the co-authors.

(2) Issam Boudraa contributed to the preparation and organization of data, literature research, and revision and editing of the manuscript.

(3) Smaïne. Chellat contributed to the scientific review, proofreading, and final checking of the manuscript.

(4) Brahim. Kebabi supervised the laboratory analyses using X-ray fluorescence (XRF) and contributed to data processing and the determination of geochemical background values, which were used as a reference for comparing and interpreting element concentrations in the samples.

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تحليل حمأة الصرف الصحي والرواسب كمؤشر للتلوث الإقليمي بالعناصر النزرة المعدنية: دراسة حالة لوادي الحراش، الجزائر

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تاريخ النشر الإلكتروني: 01 تشرين الأول 2026

الملخص

تهدف هذه الدراسة إلى تقييم مستوى التلوث في منطقة وادي الحراش (شمال الجزائر) من خلال تحليل تراكيز العناصر النزرة المعدنية والمعادن الثقيلة في حمأة مياه الصرف الصحي والرواسب. شملت الدراسة ثلاث محطات أخذ عينات هي: المنبع، وداخل محطة تطهير المياه المستعملة ببزراكي (WWTP)، والمصب. ولتحديد التركيب المعدني والمكونات الكيميائية والملوثات، أجريت تحاليل باستخدام تقنيات متقدمة شملت حيود الأشعة السينية (XRD)، وفلورة الأشعة السينية (XRF)، ومطيافية البلازما المقترنة حثيًا مع مطياف الكتلة (ICP-MS)، ومطيافية الانبعاث الذري بالبلازما المقترنة حثيًا (ICP-AES)، بالإضافة إلى تحليل الكربون العضوي الكلي (TOC). أظهرت النتائج أن الرواسب، ولا سيما في محطة التطهير، تحتوي على تراكيز مرتفعة من المعادن الثقيلة، حيث بلغت تراكيز النحاس (Cu) 2200 جزءًا في المليون، والكروم (Cr) 1160 جزءًا في المليون، والزنك (Zn) 1590 جزءًا في المليون، والنيكل (Ni) 320 جزءًا في المليون. أما في منطقة المصب، فقد سُجلت تراكيز مرتفعة من الرصاص (Pb) بلغت 330 جزءًا في المليون، والزرنيخ (As) 60 جزءًا في المليون. في المقابل، أظهرت عينات المنبع مستويات أقل من التلوث، حيث بلغت تراكيز النحاس 410 أجزاء في المليون، والكروم 680 جزءًا في المليون، والنيكل 100 جزء في المليون. كما بينت مؤشرات تقييم التلوث، بما في ذلك معامل التلوث (CF) ومعامل الإثراء (EF)، وجود مستويات مرتفعة جدًا من التلوث، خاصة بعناصر النحاس والزنك والكروم والزرنيخ، وهو ما يُعزى أساسًا إلى تصريف المخلفات الصناعية وتأثير محطة تطهير المياه المستعملة. وتؤكد هذه النتائج ضرورة الاستمرار في برامج الرصد البيئي، إلى جانب تنفيذ استراتيجيات فعالة للحد من التلوث وإعادة تأهيل البيئة، بهدف تقليل المخاطر البيئية والصحية التي تهدد منطقة وادي الحراش.

الكلمات المفتاحية:

المعادن الثقيلة، التلوث، وادي الحراش، التصريفات الصناعية، التلوث البيئي.

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