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ASSESSMENT OF THE SEDIMENT CONTAMINATION BY HEAVY METALS OF THE CHELIFF RIVER, ALGERIA

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SUMMARY

This study deals with the pollution of the greatest river in Algeria, and, therefore, 180 sediment samples from the Cheliff River from three industrial areas in Tiaret, Relizane and Chlef were analyzed. The concentration of heavy metals was determined by Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES). The results show that the sediment samples were highly polluted, particularly near Chlef and Relizane cities, with Cd (global mean: 5.48 ± 0.27 mg Kg⁻¹ dry weight), Pb (384 ± 19), Zn (1264 ± 63), Cu (442 ± 22) and Cr (399 ± 20). These results clearly exceed the ERL values recommended by US EPA. Due to natural dilution and diffusion, the level of contamination decreases near Tiaret. A comparison with the data of different sites throughout the world, the background values (Shale) and sediment quality guidelines was established. This study reveals that river sediments of Cheliff River are highly contaminated by five heavy metals: Cd, Pb, Zn, Cu and Cr.

KEYWORDS: The Cheliff River, heavy metal, industrial effluent, river sediment, contaminant, statistical analysis.

INTRODUCTION

Many river basins in the world are seriously polluted by heavy metals released to the environment from present-day and historic mining activities [1-2].

The Cheliff is the greatest river in Algeria with a drainage area of around 43,750 km² [3]. It receives wastewater from several industries (siderurgy, rubber, pharmaceutical and tannery) and urban sewages without any treatment from Relizane, Chlef and Tiaret. More than 80 % of near-by industries use the river water, and 80 % of the water from neighboring conglomerate uses and 100 % of the wastewater are rejected into the Cheliff river. Thus, the main source of sediment pollution comes from the domes-

tic sewage of the inhabitants and industrial effluents. However, the increase of population and industrial development led to a severe deterioration of water's quality [4-6], and, consequently, of the marine sediments [7].

Sediments in river react as traps for pollutant elements, especially for trace elements. Also, they can be a source of contaminants [8-11]. Rock and soil erosion constitute a major process at the earth's surface [12]. In many river watersheds, several studies have shown that the sediment load (suspended particular matter and sediments) can be significant in the transport of pollutant elements [5, 13, 14]. Sediments in rivers have multiple sources. The primary sources are neighboring manufacture and near agglomeration areas [15-17]. Several works carried out in Europe [15, 17, 18], Asia [16], North America [19], and South America [7] have studied the distribution of heavy metals in sediments, estuaries, wildlife, water, and suspended particulate matter.

No investigation relating to the pollution of the greatest river of Algeria was reported. This paper presents the early results of Cheliff river contamination by the wastewater of a mining spill. It shows the distribution of the heavy metals in river sediments with regard to their chemical behavior, the concentrations of Cd, Pb, Zn, Cu and Cr at the most sites and the changes over the 180 sediment samples. Subsequently, a correlation was established between the sites, the season and the contamination. This correlation was relied to the phenomenon of heavy metal input in the sediments. Description of these variations is beyond the scope of this paper.

SAMPLING AND ANALYSIS

Sampling Location

Nine sampling sites were chosen at the Cheliff River, along a 244 Km long profile (Fig. 1) [3]. Samples of superficial sediments were collected in July, October, Janu-

ary and April. The sites' distribution is as follows: Relizane: A, B and C; Tiaret: D, E, and F; and Chlef: G, H and I. Each site is composed of five sampling points, and at each one of them three times samples were collected. One-hundred-and-eighty sediment samples from the Cheliff river system were analyzed, since July 14th, 2002 until

April 14th, 2003. Sub-samples were taken from the central part of the grab to avoid metal contamination from the sampler. The sampling locations were selected to provide a set of representative samples of the various distinct regions, especially with regard to the influence of the industrial activities at Relizane, Tiaret and Chlef.

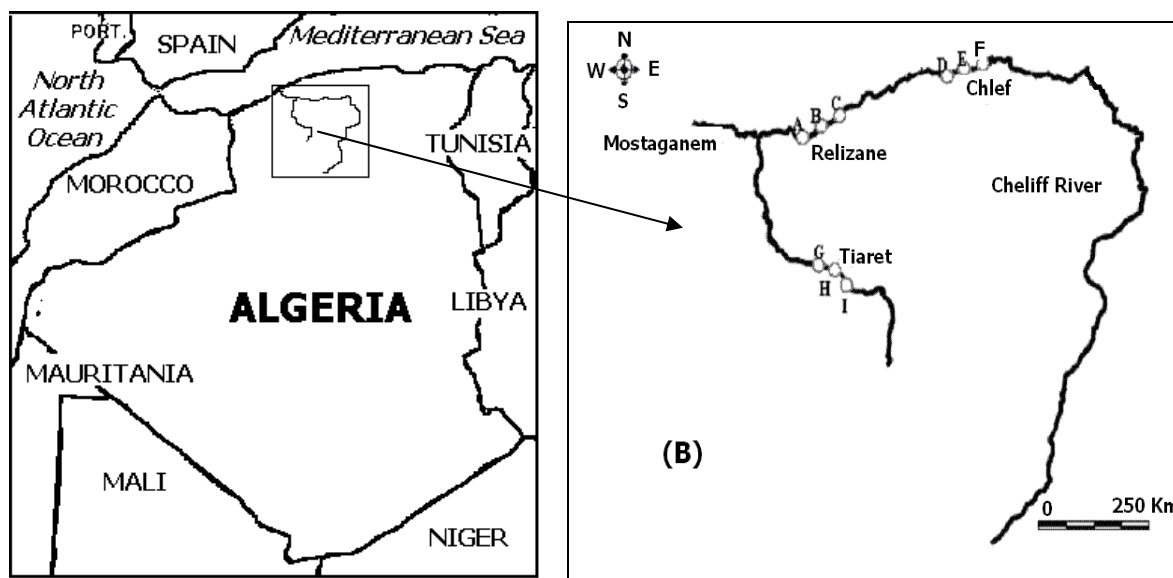


FIGURE 1 - Location of (a) the Cheliff River and (b) the nine stations A - I along the Cheliff River.

Sampling and Sample Preparation

For better protection against contamination during sampling, sediments were collected with plastic spatulas and stored in polypropylene boxes. After collection, pebbles and twigs were removed. Sediments were oven-dried at 70 °C and powdered prior to analysis [20-22]. In order to obtain representative aliquots for analyses, samples were homogenized, quartered and dry-sieved through a 65 µm mesh stainless steel sieve, without dispersion agents, to avoid contamination and modification of the samples [7, 23]. One digestion procedure and one instrumental analysis were quantitatively used to recover all metals of interest (i.e., Cd, Pb, Zn, Cu and Cr). Then, about 9 g of the mixed sample was ground in a ceramic ball mill. For recovery of the metals, the sample matrix was totally digested.

Analysis methods

Then 0.5 g of dried sample was digested with a chlorhydric, nitric and hydrofluoric acids mixture, which was dried and then diluted back to 20 ml with nitric acid. A detailed description of sample analysis methods and procedures can be found elsewhere [5, 16, 19, 24]. A sub-sample was tested for contents of cadmium (Cd), lead (Pb), zinc (Zn), copper (Cu) and chromium (Cr), with an Inductively Coupled Plasma-Atomic Emission Spectrometer (ICP-AES). All re-

sults presented in this paper are the means of triplicate analyses. The determination limit was approximately determined by inserting blind duplicates to 5% of the total number of samples analyzed. As a check of ICP-AES analytical quality of the solutions, standard reference materials (HIS-1, MESS-3 and PACS-2) were used.

Statistical Analyses of the Data

The data set obtained for the superficial sediments consisted of 20 variables (five heavy metals during four seasons) and 180 observations of samples (five samples from nine sampling points in four seasons). Pearson linear correlation matrices and factor analysis were used to determine the degree of interrelationships between the variables studied [25-27]. Multivariate analysis of these data was carried out using the STATISTICA (v.5.1) software package.

Three estimation steps were used in this study: (1) determination if the measured heavy metal concentrations were additive and normally distributed; (2) estimation of the linear correlation of pairs of the measured metal concentrations as a function of the season and direction of their separation; (3) determination of any significant differences (at $p > 0.05$) in terms of metal concentrations of sediment between different sampling sites.

RESULTS AND DISCUSSION

Results

The average concentrations, of the elements in analyzed sediments (180) are presented in Fig 2. There were large variations in the major elemental composition of the sediments, which were related to different sites and seasons. The heavy metal concentrations showed wide variation ranges, and, in many instances, were higher than back-

ground levels. The contents of heavy metals in the Cheliff River near Chlef and Relizane cities' industrial complexes present very high levels. Occasionally, the levels of chemical contamination increased again in the river sediment near Tiaret. In previous studies [1, 2, 19], sediments collected at various sites worldwide were analyzed and local enrichments of heavy metals found, but the latter could not be linked to any anthropogenic sources.

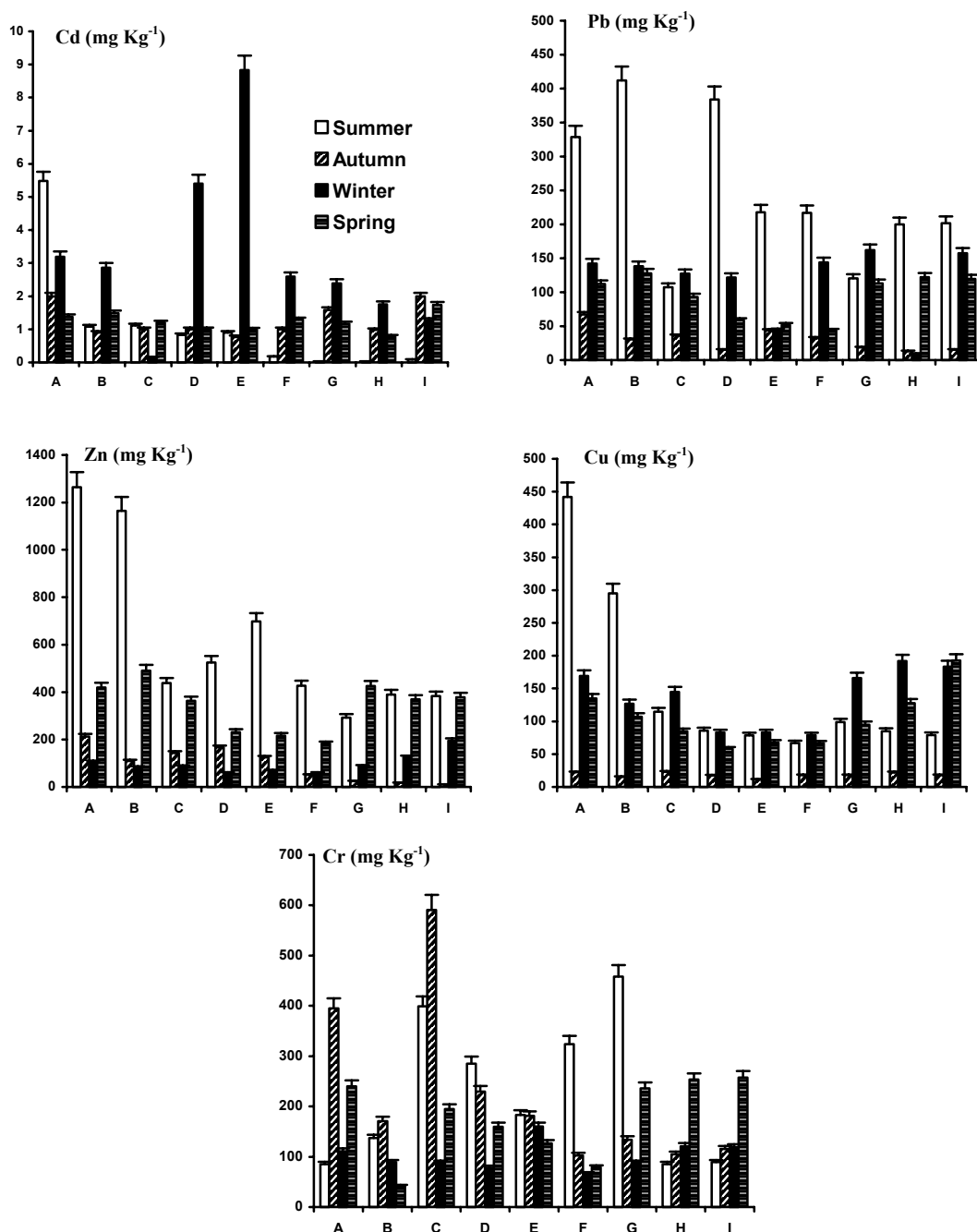


FIGURE 2 - Total heavy metal concentrations in sediments (mg Kg⁻¹, dry weight).

TABLE 1 - Minimum, maximum and average elemental concentrations (mg Kg⁻¹, dry weight) of the river sediments. Comparison of heavy metal concentrations in sediments with data published in other countries.

	Background (Shale)	ERL	Schelf Portugal	Inland Hong Kong	Seine France	Cheliff Algeria		
						Minimum value	Maximum value	Mean value
	Govindaraju [29]	Long et al. [32]	Araújo et al. [18]	Zhou et al. [16]	Clarisse [30]	(present study)		
Cd	0.14	1.2	*	4.03	2.5	0.03	8.83	1.68
Pb	31	47	30	1529	183	9.71	412	122
Zn	103	150	57	1220	360	11.2	1264	288
Cu	29	34	18	449	127	11	441	102
Cr	0.07	81	83	85.4	110	42	591	191

* no data

The systematic patterns of contaminant distributions, which were erratic, even when taking into account that they were measured at different locations and distances, cannot be identified. However, the level of contamination is closely associated with urban development and economic activities in Chlef and Relizane cities.

Table 1 summarizes average, minimum and maximum concentrations of the 5 elements determined in the sediments samples. The highest contents of Cd and Pb were found in Chlef with values of 8.83 and 581 mg Kg⁻¹ (dry weight), respectively. Occasionally, the concentrations of Zn, Cu and Cr in Relizane were three times higher than those obtained from the other sites (1264, 441 and 591 mg Kg⁻¹ dry weight, respectively). Over 98 % of the metal concentrations exceeded the Shale and the ERL values.

Statistical comparisons between river sediments according to the four seasons showed that the levels of heavy metals found in Chlef, Relizane and Tiaret were appreciably raised ($p < 0.05$).

Sediment analyses

Heavy metals relationships: In general, high concentrations of Cr, Cu, Zn and Pb were obtained in all river sediments. The magnitude of heavy metal concentrations in river sediments was $Zn > Cr > Pb > Cu > Pb > Cd$. The collected sediments from the Cheliff River were mostly polluted by these metals, because of the location in close proximity to industrial domains.

The Cheliff River significantly contained higher concentrations of Cr (7 times higher than at other sampling sites). Cu, Zn, Pb, Cd and Cr are commonly used in textile dyes, cutlery manufacture and plastic fabrication. In order to study inter-elemental associations, the Pearson's matrix correlation coefficients of the analyzed elements were computed for the total of collected sediment metal results. The correlation coefficients showing a strong elements' correlation with a single variable ($p < 0.01$) were calculated [16-11-28]. Based on the relationships between the heavy metals associated with each factor (various industrial places and seasons) and their relative importance along the Cheliff River, the sediment distribution pattern

can supply the information about the affinity of the contaminants in the sediments. The correlations between any pair of Cu, Zn, Pb, Cd and Cr are shown in Table 2. Close correlations ($p < 0.01$) were found, for Cd.su-Cu.su, Cd.su-Pb.au, Zn.su-Cu.su, Zn.wi-Cu.sp, Zn.sp-Pb.sp, Cu.wi-Cu.sp and Cu.wi-Pb.sp in the river sediments, suggesting that these metals were derived from a common origin. Metal content in river sediments is a good indicator of their usage in local industries. Remarkable increases of Zn, Cu, Pb and Cr were observed in river sediment samples collected from the Cheliff River near Relizane from July 2002 to April 2003. The results of the present study were in line with the report that the Cheliff River near Chlef's industrial site was the most polluted area, followed by Relizane and Tiaret industries. Compared to other sites in European and Asian countries [16, 18, 22, 28, 29], the heavy metal contents obtained in all sites in the present study were rather high (Table 1).

Factor analysis: In order to study inter-elemental associations, the correlation coefficients of the elements analyzed are computed for all sediment samples. The total concentration data are also subjected to factor analysis, to identify river sediment sources. At first, we establish the heavy metal distribution patterns along the Cheliff River at the industrial areas of Chlef, Relizane and Tiaret.

Previous studies have proven that factor analyses are valuable tools to recognize relationships between heavy metals, sites and the seasons, with the same behavior in the environmental studies [1, 22, 30].

The results of factor analysis based on three most significant factors indicate that they explain about 72 % of the total sample variance. The variance explanations of the factors are 31.8 % for factor 1, 25.5 % for factor 2 and 14.8 % for factor 3. On one hand, Table 3 shows that the variables of Cu.wi (total concentration of Cu found in river sediment collected in winter), Cu.sp, Pb.sp, Zn.wi, Zn.sp and Cd.au have high positive loadings on factor 1, respectively. On the other hand, the variables of Cu.su, Cd.su, Zn.su, Pb.au and Zn.au have high positive loadings on factor 2, respectively.

The factor loading of Cr.su, Pb.wi and Cr.wi shows high positive and negative values on factor 3, respectively. Three factors with different factor loadings indicate that three different contributions are involved in determining the heavy metal concentrations of river sediments. The Cu.su and Cd.su have the highest factor loadings on factor 2, respectively. Cu.su is introduced into the river by industrial wastewaters and, consequently, in sediments. Cu.wi depends obviously on Cu.su, and this by climatologic factor, precipitation during rain period, and dissolution of Cu.su fixed during dry season on sediment. In this study, it is considered that Cu.wi mainly comes from industrial effluents.

Hence, factors 1 and 2 are assumed to be indicative of the contamination sources.

The group of factors 2 and 3, Cu.su, Cd.su, Zn.su, Pb.au, Zn.au, Cr.su and Cr.wi, are largely influenced by the pollution sources of the industrial areas and climatic variations of rain period, temperature and water level during the rain time [6]. Such factor loadings indicate that both industrial wastewaters and climatic conditions contribute to the distribution of these heavy metals in the sediments along the Cheliff River in the different seasons.

TABLE 2 - Pearson correlation matrix showing the relationships between heavy metal of the river sediments samples of different season.

	Cd				Zn				Cu				Cr				Pb			
	Su	Wi	Au	Sp	Su	Wi	Au	Sp	Su	Wi	Au	Sp	Su	Wi	Au	Sp	Su	Wi	Au	Sp
Cd																				
Su	1.000																			
Wi	0.089	1.000																		
Au	0.432	-0.360	1.000																	
Sp	0.184	-0.372	0.602	1.000																
Zn																				
Su	0.786	0.241	0.106	0.287	1.000															
Wi	-0.052	-0.463	0.667	0.500	-0.150	1.000														
Au	0.778	0.356	-0.056	-0.067	0.674	-0.457	1.000													
Sp	0.261	-0.522	0.429	0.396	0.384	0.432	-0.054	1.000												
Cu																				
Su	0.886	-0.061	0.389	0.350	0.914	0.04	0.612	0.557	1.000											
Wi	0.133	-0.647	0.626	0.207	-0.057	0.775	-0.343	0.743	0.241	1.000										
Au	0.329	-0.756	0.357	-0.048	-0.088	0.266	0.082	0.325	0.249	0.607	1.000									
Sp	0.143	-0.482	0.741	0.593	0.084	0.963	-0.345	0.578	0.256	0.811	0.300	1.000								
Cr																				
Su	-0.340	-0.131	-0.221	-0.208	-0.512	-0.534	-0.058	-0.218	-0.414	-0.311	0.039	-0.616	1.000							
Wi	0.115	0.501	0.043	-0.169	0.137	0.342	-0.05	-0.018	-0.03	0.193	-0.332	0.300	-0.555	1.000						
Au	0.534	-0.237	0.038	0.013	0.248	-0.168	0.690	0.160	0.362	0.063	0.515	-0.137	0.239	-0.107	1.000					
Sp	0.144	-0.356	0.661	-0.066	-0.333	0.649	-0.190	0.277	-0.032	0.780	0.586	0.583	-0.114	0.281	0.144	1.000				
Pb																				
Su	0.391	0.350	-0.093	0.153	0.718	-0.211	0.505	0.085	0.542	-0.321	-0.264	-0.066	-0.485	-0.130	-0.128	-0.467	1.000			
Wi	0.170	-0.380	0.541	0.756	0.099	0.065	0.079	0.265	0.263	0.002	0.079	0.148	0.369	-0.619	0.159	-0.074	0.078	1.000		
Au	0.868	0.232	0.192	0.159	0.726	-0.245	0.714	0.051	0.729	-0.114	0.094	-0.071	-0.186	0.221	0.533	-0.117	0.177	0.097	1.000	
Sp	0.145	-0.583	0.482	0.344	0.225	0.641	-0.241	0.949	0.435	0.881	0.396	0.744	-0.366	0.085	0.015	0.450	0.009	0.113	-0.114	1.000

TABLE 3 - Equamax Factor analysis matrix of chemical constituents and factor scores of river sediments (n=180) and percentage of variance, explained for the individual factor loadings.

	Factor 1	Factor 2	Factor 3
Cd.su	0.149	0.924	0.050
Cd.wi	-0.640	0.172	-0.631
Cd.au	0.755	0.230	0.156
Cd.sp	0.477	0.284	0.176
Zn.su	0.010	0.939	-0.227
Zn.wi	0.900	-0.183	-0.222
Zn.au	-0.356	0.823	0.112
Zn.sp	0.720	0.307	0.131
Cu.su	0.277	0.929	0.029
Cu.wi	0.946	-0.056	-0.046
Cu.au	0.495	0.101	0.563
Cu.sp	0.946	0.036	-0.208
Cr.su	-0.427	-0.355	0.733
Cr.wi	0.173	0.042	-0.802
Cr.au	-0.019	0.491	0.531
Cr.sp	0.703	-0.184	0.062
Pb.su	-0.198	0.6094	-0.304
Pb.wi	0.179	0.226	0.650
Pb.au	-0.104	0.838	0.016
Pb.sp	0.862	0.128	0.001
Var (%)	31.751	25.489	14.802

CONCLUSION

The evaluation method of heavy metals by ICP-AES in this study allows a better sensitivity and quality of analysis of the collected sediments of the Chelif River. The data obtained suggest that contaminant forms of Zn, Cu, Cr and Pb are the most widespread in the sediments studied. The river analyzed sediments are highly polluted near Chlef and Relizane, compared to those collected from Tiaret, and are appreciably influenced by the climatic conditions. It was found that metals present different accumulation in river sediment in combined form: Cd-Cu, Cd-Pb, Zn-Cu, Zn-Pb, and Cu-Pb. Significant correlations between the various chemical heavy metal species in the sediments of the river for the four seasons were obtained: Cd.su-Cu.su, Cd.su-Pb.au, Zn.su-Cu.su, Zn.wi-Cu.sp, Zn.sp-Pb.sp, Cu.wi-Cu.sp and Cu.wi-Pb.sp. The chemical composition of the Chelif River sediments can be largely explained by transport and mixing of the different source inputs. The concentrations of Pb, Cr, Zn, Cu and Cd changed by place and in the place by a dissipation phenomenon. The normalized heavy metal distribution in the sediments provides evidence for the release of these elements into the marine environment. In fact, an enrichment of the heavy metals is detected in the river sediments in the rain season. Heavy metals may cause major adverse effects.

According to the analytical methods and the correlation studies, the sources of contamination are closely related to human activities. High concentrations of heavy metals measured in the river sediments are inter-associated and indicate that Chelif River contamination presents a public menace, which is necessary to deal with.

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