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Geochemistry of major and trace elements in sediments of Ghazaouet Bay (western Algeria): An assessment of metal pollution

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ABSTRACT

Trace metals contaminations which impact littoral ecosystems result mainly from human activities (industrial, agricultural, or urban). Thus, the particles exported and accumulated in the coastal sediments are accurate anthropic pressure gauges on this kind of environment. Spatial variations of major and trace elements concentrations in the sediments of Ghazaouet Bay (Western coast of Algeria) have been monitored between July 2010 and June 2011. Contrasted gradients of trace metals concentrations are highlighted between the preserved zones and the strongly impacted areas. The geo-accumulation index (I_{geo}) and the enrichment factors (*EF*) show that the sediments in this coastal zone are highly polluted, especially by Zn, Cd, Cu, and Pb.

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1. Introduction

Chemical elements naturally occur in the Earth's crust. However, human activities have introduced high loads of these constituents in the environment, making it now sometimes difficult to differentiate between natural and anthropic contributions.

Trace metals (TM) such as cadmium (Cd) and lead (Pb) are considered among the most critical pollutants in natural environments and for human life (Manahan, 2000) due to their toxicity, persistence, and bio-accumulation capacity at different levels of the food chain.

Trace metals are ubiquitous in the global environment (Benguedda et al., 2011; Bilos et al., 2001; Hamdoun et al., 2015; Hlavay et al., 2001; Hosono et al., 2010; Kouidri et al., 2016; Okbah et al., 2014; Rahman et al., 2014; see also the references in Chabaux et al., 2015). In natural aquatic ecosystems, TM mostly occur at low concentrations. However, in recent times, there have been rising problems with increasing contamination levels of heavy metals in those environments and pollution of aquatic habitats. Sediments in estuarine environments or on the continental shelf usually act as a sink for land-derived aquatic pollutants, and especially for trace metals (Tang et al., 2010). Consequently, sediment-associated contaminants can further influence the concentrations of TM in both the water column and biota if they are desorbed or become available to benthic organisms (Díaz-de Alba et al., 2011).

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Therefore, sediments often served as indicators of the burden of heavy metals in coastal environments as they are ultimately stored in this material (Gargouri et al., 2011).

In some works, chemical analyses in vertical sediment profiles are used to reconstruct the contamination history of aquatic systems (Evenset et al., 2007; Lepland et al., 2010; Lourino-Cabana et al., 2011). The present study aims to determine the levels of major (Al, Fe, Mg, Ti) and trace elements concentrations (V, Cr, Mn, Co, Ni, Cu, Zn, Mo, Cd, and Pb) in sediments from the coastal areas of the Ghazaouet Bay (northwestern Algeria). This area has been identified as a hot spot of coastal anthropization due to important industrial activities (Belhadj, 2008). Thus, it is now crucial to evaluate the importance of pollution according to objective criteria such as *EF* or I_{geo}. Three selected sites were chosen along the near coast to elucidate the relationship between the location of the industrial plant of zinc electrolysis as well as other polluting sources in the Ghazaouet area and the resulting heavy metal contamination in sediments.

2. Materials and methods

2.1. Study area

The Ghazaouet Bay is located in northwestern Algeria (N 37°25′19″, W 122°05′06″) at approximately 10 km from the Moroccan border (Fig. 1). The Ghazaouet town hosts an important harbor on the coast for the entire northwestern region of Algeria. Coastal seawaters are continuously exposed to industrial, urban, and agricultural wastes, releasing huge quantities of contaminants and trace metals, especially Zn and Cd, originating from a large industrial complex of zinc electrolysis, near the harbor of Ghazaouet (Benguedda et al., 2011). Moreover, tailings disposal or residues from ore exploitation by the industry occur outdoors as cliffs and are submitted to the leaching of meteoric waters that further reach the shore.

2.2. Sediment sampling

Sampling was carried out monthly between July 2010 and June 2011. Three stations were investigated in this area: the first on the beach of Wadi Abdella (A) located west of Ghazaouet. The second station is situated on the beach of Wadi Ghazouana (B) 1 km east from the first station. The third one is located inside the harbor of Ghazaouet (C), 500 m east from the Ghazaouet town (Fig. 1). Surface sediment samples were taken in shallow water. Cores are 5 cm in depth. Sediments were collected by diving in harbor "C" and by scraping surface sediments using a plastic shovel in stations "A" and "B". After collection, the samples were placed in polyethylene bags and transported to the laboratory in iceboxes. Thirty-five sediment samples have been collected for analysis.

2.3. Sediment geochemistry analysis

Sediment samples were air-dried for one week, and after performing grain size studies the finest fraction ($< 63\ \mu m$) was separated. A mixture of acids (HF, HCl, HNO_3) in proportion 1:3:1 (v:v:v), was used for the sediment digestion in a microwave oven (Anton Paar Multiwave 3000). Acids exhibiting high purity were employed. Extracts were then evaporated to dryness and diluted afterwards with Milli-Q water. The resulting solutions were stored at 4 °C until ICP-MS quantitative analysis of trace elements (Al, Fe, Mg, Ti, V, Cr, Mn, Co, Ni, Cu, Zn, Mo, Cd, and Pb) using an Agilent 7700X instrument. The accuracy of the analytical procedures for the determination of total metal concentrations was checked using the NIST 1646a estuarine sediment standard. These were analyzed under the same experimental conditions. Two replicate analyses of this reference material processed in the same time as samples are in good accordance with certified values (Supplementary material, Table S1).

2.4. Geo-accumulation index (I_{geo})

According to Müller (1969), the geo-accumulation index (I_{geo}), highlights the assessment of sediment and soil pollution by heavy metals (e.g., Amin et al., 2009; Singh et al., 2005; Williams and Block, 2015). The values of the geo-accumulation index can be calculated according to Müller (1969) as follows:

$$I_{geo} = \log_2(C_n/1.5\,B_n) \quad (1)$$

where C_n is the measured concentration of the examined metal (n) in the examined bottom sediment, and B_n the geochemical background value of element n in the surrounding rocks or, if not available, in the average shale (e.g., Dali-youcef et al., 2005; Turekian and Wedepohl, 1961); 1.5 is the correction factor addressed due to lithogenic effects (Lin et al., 2008).

2.5. Enrichment factor (EF)

It is common to estimate the anthropogenic impact on coastal sediments via the calculation of trace element enrichment factors (*EF*) (e.g., Kouidri et al., 2016; Lourino-Cabana et al., 2011; Rahman et al., 2014; Rodriguez-Barroso et al., 2010). They result from a twofold normalization of the analyzed samples with reference materials that refer to uncontaminated background levels (Dickinson et al., 1996; Salomons and Förstner, 1984) – see Eq. (2). Because TM are generally associated with the finest particles, TM concentrations must be divided first by the concentrations of an element (X) in the samples that is of natural origin and that is highly associated with the grain size fraction of clays. Then, a second normalization with a reference background sample is applied. It reflects natural TM concentrations that are commonly recorded in the environment (soils or surrounding rocks).

$$EF = (TM/X)_{\text{sample}}/(TM/X)_{\text{reference}} \quad (2)$$

Theoretically, if the *EF* is higher than one, it may be indicative that the sediment is mainly originating from an anthropogenic source and can be used for assessing the degree of pollution. In fact, it is more commonly admitted that $EF > 2$ reflects enrichment from anthropogenic

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