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Trace element fluxes during the last 100 years in sediment near a nuclear power plant

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ABSTRACT

The Salada coastal lagoon is located in Veracruz (Mexico) near the Laguna Verde Nuclear Power Plant (LVNPP). Currently, the lagoon receives the cooling waters used in the LVNPP. To evaluate the fluxes and mobilization of trace elements due to human activities in the area, two sediment cores from the coastal flood plains of Salada Lagoon were analysed. Cores were collected using PVC tubes. Sediments cores were analysed every centimetre for dating (²¹⁰Pb by alpha detector) and trace metal analysis using ICP-Mass Spectrometry. The dating of both sediment cores covers the period from 1900 to 2013, which includes the construction of the LVNPP (1970's). The Normalized Enrichment Factor shows enrichment of Ag, As and Cr in both sediment cores. These enrichments correspond to the extent of mining activity (which reached a maximum in the 1900's) and to the geological setting of the coastal zone. The profiles of the element fluxes in both sediment cores reflected the construction and operation of the LVNPP; however, the elements content did not show evidence of pollution coming from the LVNPP.

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1. Introduction

Anthropogenic activities release large quantities of trace elements to the environment. The energy production and mining industries may release trace elements which can accumulate in water, sediment, soils and organisms (Chang and Page, 2000; Wuana and Okieimen, 2011).

Nuclear power plants (NPP) convert atomic energy into heat for the generation of steam to drive a steam-turbine that is connected to a generator producing electricity (GES, 1997). NPP are regulated by legislation such as clean energy programs that is focused on protecting human health and the environment; however, there is a wide variety of environmental impacts associated with power generation technologies (USEPA, 2015). Studies of sediments sampled near these industries show evidence of trace element enrichments in the vicinity of nuclear power plants in northern

Serbia and Montenegro (Cd, Cu, Cr, Zn, and Hg; Dalmacija et al., 2006), the Czech Republic (Al, As, Cd, Co, Cs, Fe, Mn, Se, V, and Zn; Thinova et al., 2014) and China, and the highest metal levels were found in samples located close to nuclear power plants, probably as a result of nuclear waste discharges (Hg, Pb, Cu, Zn, Fe, Al, Ni, Sr, Li, and Co; Gu et al., 2012).

Sediments can reflect the quality of a coastal system as well as the historical hydrogeochemistry; total trace elements concentrations in sediment cores is performed to determine element anomalies in zones of mineralization as well as from pollution sources (Salomons and Förstner, 1984). Sediments are a sink of elements subjected to a variety of physical biological and chemical processes (Chester and Jickells, 2012). Geochemical studies of trace elements in sediments may help to identify the background level, type, magnitude and provenance of environmental pollution (Jaramarini et al., 2013; Zhou et al., 2014; Páez-Osuna et al., 2015); fluxes (Ruiz-Fernández et al., 2004); and the periods of environmental pollution (Emadodin, 2011; Ontiveros-Cuadras et al., 2014; Zhang et al., 2014). In previous studies, researchers were able to distinguish between the influences of industry, mining, and population

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centres in their study areas. Trace elements such as: Cu, Hg, Ni, Sr, V, and Zn, are used as indicators of pollution coming from the industrial discharges of nuclear power plants (Bojakowska and Krasuska, 2014). On the other hand, Ag, As, Cd, Co, Cr, Cu, Hg, Pb, V, and Zn have been used to identify mining activities (Marmolejo-Rodríguez et al., 2011; Sánchez-Martínez et al., 2013; Horák and Hejzman, 2013).

Due to the scarce studies on trace elements in sediments near nuclear power plants, two sediment cores were collected in Salada Lagoon in order to: i) estimate the origin and possible enrichments of some major and trace elements, ii) evaluate the fluxes of the enriched elements during the last 100 years, iii) assess whether there is any influence of the Laguna Verde Nuclear Power Plant (LVNPP) on the sediments of the lagoon.

2. Study area

The Salada Lagoon is a coastal system located in the central part of Veracruz State (Mexico) between 19° 42' and 19° 43' N; 96° 24' and 96° 25' W, within the Actopan La Antigua hydrological region (INECC, 2014). The lagoon is triangular, with a 51 ha surface area; the climate is warm sub-humid with summer rain, and the mean annual temperature varies from 21 to 28 °C (CONABIO, 2012). It is a shallow system (maximum depth of 3 m) that initially had transient communication with the sea through a narrow sand barrier that caused periods of seasonal drought in the lagoon (Lankford, 1977; CFE, 1989). The lagoon is microtide influenced (fluctuations between 0.5 and 0.7 m, with the highest tides during October and November, and the lowest between June and July; SEMAR, 2016) and receives no direct fresh water discharge, only rainwater runoff. In fact, salinities of up to 70 PSU were recorded in the lagoon between the years 1978–1979.

The Salada Lagoon currently receives the coolant water discharge from the LVNPP, located ~0.2 km distant (Fig. 1), which was built between 1976 and 1995 (Morales-Lozano, 2011). In 1981, the cooling water discharge canal of the LVNPP was built through the eastern side of the lagoon. The opening of the water discharge channel established a permanent communication between the lagoon and the sea, allowing values of salinity of up to 35 PSU in recent years (Rodríguez-Castañeda, 1994). Most of the coastal zone of Veracruz State is composed of Quaternary alluvium deposits, and in the west of the Lagoon, there is a predominance of Tertiary andesites intercalated with basaltic breccia (Fig. 1; SGM, 2014a,b).

Moreover, there are several mining regions in Veracruz State (viz. Huayacocotla, Chiconquiaco, Zongolica-Atoyac, Los Tuxtlas and Cuenca Salina); among them, the Chiconquiaco (silver mining) region is the closest to the Salada Lagoon (Fig. 1; SGM, 2014a,b). The dominant winds in winter in Veracruz State are the “Nortes” (a cold northeasterly wind that blows in Mexico and on the shores of the Gulf of Mexico) coming from the polar zones (Llanos-Arias and Cervantes-Pérez, 1995; Oliver, 2005). During summer, the wind turns toward the north in the northwestern Gulf due to the influence of the North Atlantic semi-permanent atmospheric high-pressure system over the northeastern region (Zavala-Hidalgo et al., 2014). In addition to the presence of trace elements related to mining activity, there were also high concentrations of As and Cr in sediments of the coastal zone adjacent to Veracruz state (Celis-Hernández et al., 2013); the authors attribute these high concentrations to the local lithology.

3. Materials and methods

3.1. Sampling

Salada I and Salada II (SAI and SAII) sediment cores taken at 19°

42' 47.2" N, 96° 24' 47.8" W, and 19° 42' 45.1" N, 96° 24' 42.0" W, respectively (Fig. 1). The cores were sampled in February 2013 at 743 m from the LVNPP, with a distance of 180.5 m between them. The SAI site is further from the channel with respect to SAII, which is located next to the lagoon channel. The sampling location were chosen base on the hydrodynamics of the lagoon, SA-I reflect processes occurring on calmer conditions than SA-II.

The shores of the Salada Lagoon are coastal flood plains zones. These zones have a bare sediment that is only flooded at the highest tides. The cores were taken from the coastal flood plains of the lagoon. Within approximately 5 m, there is mangrove vegetation (*Avicennia germinans*, *Rhizophora mangle*, *Laguncularia racemosa*, among others; Rodríguez-Castañeda, 1994). Because these zones are inundated by the tides, sediment cores were collected during the lowest tide, therefore, there was no interstitial water at the surface of the core.

The cores were sampled manually in a flood plain using polyvinyl chloride (PVC) tubes (50 cm in length and 12 cm in diameter) cut to a 1 cm contiguous sections down core. Sediments were freeze-dried, powdered in an agate mortar, and finally stored in plastic bags until analysis.

3.2. Sediment characterization

Grain size analysis was performed by the laser diffraction technique with a Malvern Masterizer Hydro 200MU device. Magnetic susceptibility (MS) was measured with a Bartington MS2 sensor, the results expressed in SI units, and reference materials were determined to assess the quality of the results. Organic carbon content (C_{org}), analysis was performed by an elemental analyser (model COSTECH 4010); the reference material was also analysed.

3.3. Dating with ^{210}Pb and ^{137}Cs

Total ^{210}Pb ($^{210}\text{Pb}_{tot}$) analyses were carried out reaching an 18 cm depth of the sediment core ($n = 18$) by alpha spectrometry (EG&G Ortec TM silicon detector, model 574) through ^{210}Po , assuming secular equilibrium between both radioisotopes (Flynn, 1968; Ruiz-Fernández et al., 2009). The accuracy of the ^{210}Po method was 99% and the analysis of replicates yielded a variation coefficient of 9.5%. The calculation of the ^{210}Pb atmospheric flux was carried out by multiplying the total inventory of ^{210}Pb (Bq cm^2) by the radioactive decay constant of ^{210}Pb (Appleby and Oldfield, 1992). ^{137}Cs activity was measured using gamma ray spectrometry with a HPGe well detector (Ruiz-Fernández et al., 2009).

3.4. Major and trace element analysis

Major (Al, Fe, K, Mg) and trace elements (Ag, As, Be, Ca, Cd, Co, Cr, Cu, Hg, Li, Mn, Mo, Na, Ni, Pb, Sr, U, V, Zn) were determined using ICP/MS (Perkin Elmer Sciex ELAN 9000), the sediment was acid digested (HF, HClO_4 , HNO_3 , HCl) in Actlabs laboratories (Ontario, Canada). Validation of the analysis was performed using different certificated reference materials (PACS-2, MESS-3 and AGV-2) and recovery percentages above 75% were obtained for all of the elements. The variation coefficient of trace element analysis were <5%. The details of the validation methods are presented in Table 1.

3.5. Normalized enrichment factors (NEF) calculation

To compare the results of the element enrichments in the sediment, NEF calculations were performed, considering the elemental composition in the upper continental crust (UCC; Wedepohl, 1995) and the regional background levels (BL, Table 2). The NEFs were normalized with respect to aluminum, as is

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