



The mobility and speciation of lead and cadmium in Bahr El Baqar region, Egypt

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ABSTRACT

Under continuous irrigation with Bahr El Baqar drain water in Egypt, chemical speciation of Pb and Cd in soils is critical to understand, their mobility and potential toxic effects. In the present study, sorption isotherm experiments of these metals in nine representative soils were assessed at varying metal concentrations (20, 40, 60, 80 and 100 mg l⁻¹), solid to liquid ratio 5 g/50 ml and contact time (24 h). Additionally, the kinetic experiments were performed using initial concentration of 100 mg l⁻¹ Pb or Cd and contact time of 2, 4, 8, 12 or 24 h. The two experiments were carried out at 298 K. Low amounts of Pb were found in exchangeable and carbonate fractions comparing with the residual, oxides and organic fractions for the most studied soils. On the other hand, the residual fraction of Cd speciation was dominant for all the studied soils, followed by carbonate fraction. Both Langmuir and Freundlich models isotherm for Pb and Cd adsorption by these soils were fitted. The second-order kinetic model was the best fit model for the adsorption mechanism in these soils. These soils are still able to be retained and more loaded with Pb and Cd, since they did not reach their saturation capacities. Taken together, continuous accumulation of these toxic metals will be exposed to the environment to real disaster under bad management in this region.

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Introduction

The agriculture in Egypt is mostly dependent on fresh water, which is becoming a limited source due to increased population, shrinking capacity of limited surface water reservoirs and poor-managed distribution system of irrigation water. The use of raw drainage effluent such as Bahr El Baqar drain for agricultural crop production has become prerequisite, especially on urban agricultural lands with water shortage. Reusing this drainage water may cause adverse effects on soil, crop, animal, and human health, since Bahr El Baqar drain is considered as one of the most polluted drains in Egypt [1–4].

Four main sources of pollutants which cause deterioration in Bahr El Baqar drain are: the industrial activities in Shoubra El-Khema including metal production, food processing, detergents and soaps manufacturing, textile finishing and paper production; industrial activities in Zagazig area; wastewater discharged from Belbeis drain to Bahr El Baqar; and domestic discharge received from rural areas around Bahr El Baqar drain, the discharge of industrial, agricultural and municipal wastewaters in Bahr El Baqar drain led to contamination of the surrounding with many kinds of pollutants, especially heavy metals such as lead, and cadmium

[4,5]. Heavy metals are detrimental to the environment because of their non-biodegradable and persistent nature. Metals such as cadmium, mercury and arsenic are the most hazardous [6]. The continuous use of such effluent led to the accumulation of heavy metals in agricultural soils due to their high retention capacities. Among these heavy metals, Pb and Cd are getting increased concern because they are two of the most ecotoxic heavy metals.

The toxicity of Pb and Cd is enhanced through accumulation in living tissues and consequent biomagnifications in the food chain [7]. Lead has an environmental toxicity as the metal releases into air and water from a multitude of sources such as Pb smelter, battery manufacturer, paper and pulp industry, boat and ship fuel and ammunition industry [8]. In particular, Pb has deleterious effects on the nervous system, blood circulation system, cardiovascular system, vital organs (brain and the kidneys), and restricted development of intelligent quotient (IQ) [9]. However, toxicity of Cd has received as much attention as those of Pb. Metal plating, smelting and mining industries, cadmium-nickel battery manufacture, phosphate fertilizers, paints and pigments manufacture, and alloy industries are main sources of Cd in the environment [10]. Cadmium poisoning leads to lung, liver and kidney damage, bone lesions, cancer and hypertension, and the dreaded itai-itai diseases [11]. Thus, Pb and Cd were representative as an indicator on the pollution degree in Bahr El Baqar region. The dynamic equilibrium among the different fractions of the metal determines its mobility and bioavailability in the soils. The pH, Eh

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and amount and type of soil colloids (organic matter, clays and oxides) are the most important edaphic factors that control concentrations [12].

In the absence of review of literatures on chemical speciation and sorption studies of Pb and Cd in the soils of Bahr El Baqar region, therefore objectives of this study were to; investigate the fate and behavior of Pb and Cd in the soils of this region, the relation between the components of these soils with the chemical speciation of the studied metals and then assess the status of the pollution in Bahr El Baqar soils and whether these soils are able to be loaded with other levels of these metals under continuous irrigation by Bahr El Baqar drain by which; (1) determine the components of the studied soils and the type of clay minerals by X-ray fluorescence and X-ray diffraction analyses, (2) perform the chemical fractions of Pb and Cd which have the major effects on their movement in soils by sequential extraction, (3) doing the correlation between the selected physicochemical properties of these soils with the major fraction of the studied metals and (4) generate sorption isotherms and kinetic sorption studies by batch equilibrium experiments for the predication of sorption capacities of soils. Finally, helping the decision maker in putting good solutions in management of this region and then decrease size of the pollution problem in which.

Materials and methods

Collection and analyses of soils

The study area is located in northern Egypt, Bahr El Baqar region, between 32°05' and 32°16' longitude and 30°56' to 31°07' latitude. Nine soil samples were collected, air-dried and crushed with a wooden mortar and then passed through a 0.5 mm mesh-screen. The electrical conductivity (EC) of soils was determined in soil extract (1:2 ratio of soil to water). Particle size distribution was determined by the pipette method [13]. Soils were also characterized for their carbonate content according to the method described by Allison and Moodie [14]. Soil pH was measured in deionized water (pH_w) and in 0.01 M CaCl₂ (pH_{CaCl₂}) (in 1:2.5 suspension), and organic matter (OM) was determined by wet digestion [15]. Soil cation exchange capacity (CEC) was determined by Na-method [16].

X-ray diffraction (XRD) analysis

The chemical composition of the studied soils was determined by X-Ray fluorescence (XRF) (Philips, PW2400) on fused beads containing Li₂B₄O₇ flux, and loss on ignition (LOI) was equal to mass loss upon firing at 1000 °C. The phase composition of soil samples was identified by XRD technique using cobalt target ($\lambda = 1.791 \text{ \AA}$) and a nickel filter. A Philips X-ray diffractometer was used at 40 kV and 20 mA. The selected scanning speed was 1° min⁻¹ and the sample was used to fill the aluminum mold of the diffractometer with an average thickness of ~1.0 mm. The phases were identified by a correlation with the corresponding standard X-RAY CARDS (ASTM) [17]. Metal contents for each treatment were measured by atomic absorption spectroscopy (Perkin-Elmer, AAS, flam, Series711838 v1.26).

Sequential extractions (localization phase speciation)

A sequential extraction scheme was established according to a modified method of Tessier [18] and Shuman [19]. This method was enabled to identify seven fractions: the exchangeable fraction (exch.), carbonate fraction (car.), manganese oxide fraction (Mn ox.), amorphous iron fraction (am. Fe), crystalline iron fraction (Cry. Fe), organic matter fraction (OM) and residual fraction (res.).

The experiments were conducted in 50-ml centrifugation tube with 0.5 g of each studied soil. A continuous agitation was maintained during the extraction time. Between each successive extraction, separation was done by centrifugation (30 min at 4000 rpm). The supernatant was removed and the residue was washed with 16 ml of deionized water. The extraction and wash supernatant were pooled. Total concentrations of Pb and Cd in these soils were determined by acid digestion [20] using quantities of concentrated HNO₃ (5 ml), HF (5 ml) and HClO₄ (1.5 ml).

Sorption isotherm experiments

Batch adsorption tests were conducted to assess the adsorption capacity of the studied soils for Pb and Cd. Metal ion solutions were prepared by diluting 1 g l⁻¹ of stock Pb or Cd ion solution, which obtained by dissolving a weight quantity of nitrate salts of these metals (Merck). Specifically, solutions of Pb and Cd were prepared separately at concentrations of 20, 40, 60, 80 and 100 mg l⁻¹ and applied to different soil samples.

The solutions were added to the soil samples with ratio of 5 g of dry soil to 50 ml of heavy metal solution in 50-ml plastic centrifuge tubes. The soil-suspension samples were equilibrated by shaking at 300 rpm for 2 h at 298 K, after that the samples were kept up to 24 h on which and centrifuged at 3000 rpm for 20 min and the supernatants were collected.

Kinetic study

Kinetic experiment was performed using mono initial metal concentration of 100 mg l⁻¹ Pb and Cd. A 5 g of each soil was placed in five plastic centrifuge tubes and mixed with 50 ml of Pb or Cd solution. The tubes were shaken on a shaker (300 rpm) for 2 h and leaved to reach the contact times (2, 4, 8, 12 and 24 h) at 298 K. Then, the suspensions were centrifuged at 3000 rpm for 20 min and the supernatants removed by filtration (Whatman No. 42). Pb and Cd were analyzed in the filtrates by atomic absorption spectroscopy.

The total amount of metal retained by the solid phases was obtained as follows:

$$q = \frac{(C_0 - C_t)V}{W} \quad (1)$$

where q is the amount of metal sorbed per unit mass of soil (mg kg⁻¹), C_0 and C_t are the metal concentration in the initial solution and after the retention time in the filtrate, respectively (mg l⁻¹), V is the volume of adding solution (l) and W is mass of the air dried soil (g).

Statistical analysis

The correlation coefficient (R^2) was done for some physico-chemical parameters in the studied soils, i.e., total carbonate% with the fraction of metal bound to carbonate (carbon.), the total MnO% with the fraction of metal bound to Mn oxide (Mn ox.), total iron as Fe₂O₃% with the fraction of metal bound with amorphous iron (am. Fe), total iron as Fe₂O₃% with the fraction of metal bound to crystalline iron oxides (cry. Fe), and OM% with the fraction of metal bound to organic matter (OM). Statistical analysis was performed using SPSS statistical package, version 16.0 (SPSS Inc., MI, USA).

Results and discussion

Characterization of the soil samples

As shown in Table 1, the soils have a range of 46.1–80% clay content and found to be textural clay soils. The pH_w values of soils

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