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Research article

Role of phosphogypsum and NPK amendments on the retention or leaching of metals in different soils

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ABSTRACT

Column leaching tests were conducted to investigate the effects of soil physicochemical characteristics on metal mobility in the subsurface. The metals investigated originated from disposed industrial waste byproducts and from agrochemicals spread over the farmlands. Soil column tests can provide insights into leaching of metals to underlying water compartments. The findings of this study can be used for prevention strategies and for setting risk assessment approaches to land-use and management, and soil and water quality and sustainability. Soils collected from an industrial (IS) watershed and an agricultural (AQ) hydrographic basin were used in soil column leaching experiments. The soil samples were characterized for mineralogy, functional groups, grain size, surface charge, soil type, porosity, and cation exchange capacity (CEC) along with elemental composition. Varying concentrations of phosphogypsum industrial waste or agrochemical (NPK fertilizer) was then added to the surface of the packed columns ($n = 28$). The columns were subjected to artificial rain over a period of 65 days. Leachates were collected and analyzed for dissolved Na^{+}, K^{+}, and Cd^{2+} throughout the experimental period, whereas residual Cd content in the subsurface soil was measured at the end of the experiment. Physicochemical characterization indicated that the AQ soil has a higher potential for metal retention due to its fine clay texture, calcareous pH, high organic matter content and CEC. Metal release was more prominent in the IS soil indicating potential contamination of the surrounding soil and water compartments. The higher metal release is attributed to soil physicochemical characteristics. High calcium concentrations of phosphogypsum origin is expected to compete for adsorbed bivalent elements, such as Cd, resulting in their release. The physicochemical characteristics of the receiving media should be taken into consideration when planning land-use in order to achieve sustainable development. Soil physiochemical characteristics play a key role in determining the behavior and fate of elements upon application of amendments. Sandy soils should not be assigned to industrial zones or landfills due to their high permeability, unlike fine clay soils. Furthermore, application of fertilizers on sandy soils can threaten groundwater quality, whereas their extensive use on clayey soil can cause soil salinisation.

1. Introduction

Mobility, transport and leaching behavior of metals and other contaminants in the environment have been of great interest to many researchers (Du Laing et al., 2009; Eggleton and Thomas, 2004; Kalnejais et al., 2010; Lesven et al., 2010; Syrovetnik et al., 2007; Thevenon et al., 2013). Soil column leaching studies, performed under controlled conditions, can help in understanding the

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behavior of certain elements or compounds in a defined area (Alvarez, 2007; Arfania and Asadzadeh, 2015; Maszkowska et al., 2013). Subsequently, such studies can allow the assessment of the effects of the various elements on fauna and flora, and their possible leachability to groundwater (Zhao et al., 2009). There are diverse factors and processes impacting metal behavior in soil and sediment columns, which in turn rely on metal characteristics and their encompassing matrices. The mobility of metals depends on the speciation, sorption behavior, specific electronegativity, ionic radius and reaction time, where mobility can further be impacted by the physicochemical properties of soils and sediments, such as pH, redox potential, cation exchange capacity (CEC), mineral composition, grain size, surface charge, soil structure and texture, permeability, organic content, and vegetation cover (Alvarez, 2007; Balkis et al., 2009; Carrillo González et al., 2006; Herndon and Brantley, 2011; Hiller et al., 2010; Maszkowska et al., 2013).

Competition for more surface areas leads to changes in land-use, unfortunately often neglecting proper management strategies, or the specificity of the receiving media (Smith et al., 2010). Excessive usage of agrochemical fertilizers and soil amendments were randomly applied on agricultural fields causing metal remobilization from the soil matrix and subsequent infiltration to groundwater (Atafar et al., 2010; Zhang and Shan, 2008). Furthermore, the demand for more landfills and industrial waste disposal sites also increased (Leão et al., 2004; Younes et al., 2015). Under weathering conditions, these industrial wastes loaded with trace metals and radioactive materials, and mostly with low pH, have a high potential to infiltrate ultimately to groundwater. Subsequently anthropogenic activities can cause an imbalance in the ecosystem by degrading soil quality, increasing soil salinity, and posing hazards to both open water bodies and groundwater qualities (Négrel et al., 2014). Soil physicochemical characteristics are usually poorly considered when dedicating a specific land area for human activities. Internal soil physicochemical properties and the inter-linkage with the watershed characteristics strongly influence the behavior of contaminants and metals in a specific soil. Thus, identifying the soil properties and addressing its characteristics will contribute to a better understanding of the behavior of a potential contaminant. By applying the aforementioned notions, implementing prevention strategies and setting risk assessment approaches to land-use and management can be improved.

The eastern Mediterranean is known for its multivariate topographies and different microclimates. The orographic formations separate the coastal plain from the inland valley, thus inducing the presence of different environmental phenomena controlled by geological formations, microclimate properties, demographic distributions, and land-use activities (Edgell, 1997; MoA (Ministry of Agriculture), 2003; USAID, 2003). Urbanism and industrial factories are mainly located in capitals and along the Mediterranean coast, while agricultural fertile plains are spread among a variety of lands (Güllü et al., 2005), as is the case of Lebanon (USAID, 2003). In this study, the problem lies within the behavior of metals affected by: (i) industry by-product phosphogypsum (PG) that is stored as stockpiles near the northern Lebanese coast, which proved to be a socio-economic stress factor on the nearby Mediterranean Sea (Ammar et al., 2013), and (ii) agrochemicals that are overused and accumulated in soils and sediments at large scales near vital water bodies (Kanbar et al., 2014). PG has high potential to release hazardous metals that consequently cause toxicity to soil microflora (Hentati et al., 2015; Jalali et al., 2016). However, PG has been used as a soil amendment as well (Alva and Sumner, 1988). Nevertheless, the potential hazards of released metals during PG dissolution can be mitigated by adjusting soil pH (Ammar et al., 2013). The present study aims to evaluate the influence of soil physicochemical characteristics on the fate of major elements (Na and K) and trace metals (Cd) derived from anthropogenic activities. This study delineates the mobility behavior of metals in soil leaching columns, and assesses the influence of soil texture, contaminant type and dosage on such solubilisation. Soil column leaching studies are a commonly used approach to risk assessment of socio-economic activities on a specific soil type. More specifically, the effects of PG and NPK fertilizer at different concentrations on metal retention or leaching are monitored by following the concentrations in the leachates of monovalent cations (Na^+ and K^+) and the trace metal (Cd) in two different Mediterranean soil types. This study can provide guidance on effective application of amendments and agrochemicals.

2. Materials and methods

2.1. Study sites

The study areas chosen in this work comprise two sites from different watersheds (Fig. 1). The first one is located in the vicinity of the Qaraaoun reservoir, which is impacted by agricultural and industrial activities in the upper Litani watershed (Kanbar et al., 2014). Soil was collected from both sides of the reservoir, homogenized and treated as a distinct soil (AQ). The second one is a site perturbed by industrial activity and environmental degradation in Northern Lebanon, mainly via chemical fertilizer industries (Ammar et al., 2013).

2.2. Disturbed soil columns

Soil column leaching experiments were carried out in the laboratory at 20 °C under controlled conditions. The soil columns were amended with products containing metallic trace elements and nutrients. The columns were subjected to artificial raining events on top of the columns for a period of 65 days to allow the assessment of the behavior of selective elements in the leachate and the subsurface of the soil columns.

A total of 28 packed columns were randomly distributed into two discrete systems; each system is composed of 14 columns designated for a soil from each watershed. The first set of packed columns contained soil collected from the industrial watershed in Selaata (IS), whereas the second set of soil was collected from an agricultural watershed in Qaraaoun (AQ) (Fig. 1). More information about the soil column leaching set is found in the supplementary material (SM1). Volumetric (θv) and gravimetric (θg) water holding capacities in the soil columns were calculated as the percentages of volume and mass of water to soil, respectively. The soil amendments, containing trace metals, selected for this study included commercial NPK 20-20-20 fertilizer (hereby referred as NPK) and phosphogypsum (PG). The first amendment was chosen for its extensive use in agriculture, whereas the other one is an industrial by-product usually deposited on soils as stockpiles near industries, as is the case in Selaata (Northern Lebanon). The PG sample was collected from a slurry in the industrial factory (Ammar et al., 2013).

Briefly, each set of 14 columns was further divided into subsets of controls ($n = 2$), columns amended with NPK 1X ($n = 4$), NPK 2X ($n = 2$), PG 1X ($n = 4$) and PG 2X ($n = 2$). For NPK amended soils, 1X and 2X contents refer to normal and double quantities of that fertilizer typically applied to a surface of soil which is equivalent to that of the columns' (7.94 mg and 15.88 mg, respectively). As for PG, the two dosages used represented saturation and double saturation (2 g/L and 4 g/L, respectively) in ultrapure water (Ammar et al., 2013). A summary of the columns nomenclature and treatments can be found in the supplementary material (SM2). At the beginning of the experiment, 70% of water holding capacity was reached