

# Impact of a Phosphate Amendment on the Environmental Availability and Phytoavailability of Cd and Pb in Moderately and Highly Carbonated Kitchen Garden Soils



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(Received February 26, 2017; revised April 8, 2017)

## ABSTRACT

The behaviour of metals mainly depends on soil pH, carbonate contents and contamination level, which should be considered for the management of contaminated soils. In this study, kitchen garden topsoils (0–25 cm) were sampled from the area around three smelters in France, with different Cd and Pb contamination levels. Effect of a phosphate amendment (a mixture of diammonium phosphate and hydroxyapatite) on the environmental availability and phytoavailability of Cd and Pb was evaluated by different chemical extractions and cultivating lettuce (*Lactuca sativa* L.), respectively. Changes in the distribution of Cd and Pb were found in most contaminated soils after phosphate amendment. An increase of Cd and Pb in the residual phase was highlighted in almost all carbonated contaminated soils, whereas a decrease of Pb in the exchangeable, water and acid-soluble phase was observed in most contaminated soils with the lowest carbonate contents. The concentrations of extractable Cd and Pb using calcium chloride and acetic and citric acids generally decreased after the soil amendment. Lettuces grown on amended soils were acceptable for human consumption as regard to Pb concentration. In contrast, some lettuces were unacceptable for human consumption, since the concentrations of Cd in the leaves were higher than the European legislation limit. Surprisingly, in carbonated soils with very low concentration of Cd, the Cd concentrations in lettuce reached up to the European legislation limit, making the lettuce unacceptable for human consumption. Our study highlighted the fact that the total metal concentration in soils does not always allow to predict the metal accumulation in the edible parts of vegetables in order to make a judgement about their acceptability or unacceptability for human consumption.

**Key Words:** chemical amendment, contamination, immobilisation, lettuce, metal, urban soil

**Citation:** Waterlot C, Pruvot C, Marot F, Douay F. 2017. Impact of a phosphate amendment on the environmental availability and phytoavailability of Cd and Pb in moderately and highly carbonated kitchen garden soils. *Pedosphere*. 27(3): 588–605.

## INTRODUCTION

The management of contaminated soils is of great interest for scientists and researchers and still remains a major concern for population and political authorities. Rehabilitation techniques depend mainly on the type of pollutants (*e.g.*, organics or inorganics), the contamination level, the surface of the contaminated area, the physicochemical parameters of soils and the soil use after rehabilitation. Among these techniques, some of them consist in separation/concentration technologies (*e.g.*, soil removal, soil flushing and electrokinetic extraction), containment and immobilization techniques (*e.g.*, using barriers and reducing permeability and solubility) and toxicity reduction technologies (*e.g.*, chemical and physicochemical treatments). All of these techniques are often expensive and may reduce biological activities and soil fertility

and cause destruction of soil structure (Smith *et al.*, 1995; Dermont *et al.*, 2008). However, soft technologies based on phytoremediation processes like phytoextraction and phytostabilisation are known to be cost effective, energetically inexpensive and environmentally sound (Cunningham *et al.*, 1995; Guerinot and Salt, 2001; Pilon-Smits, 2005). The phytoremediation techniques, from which plants are used to remove, transfer and/or stabilize inorganic pollutants in soils (Hughes *et al.*, 1997; Salt *et al.*, 1998), are generally highly acceptable to the public. They are mainly appropriate to large areas instead of small areas like private parks, lawns or kitchen gardens. Especially for contaminated kitchen garden soils, which are complex due to the presence of many pollutants (organics and inorganics) and their heterogeneity, a first method consists of removing the contaminated materials and replacing with uncontaminated soils (Hajdu and Licsko, 1999;

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Lanphear *et al.*, 2003; Nielsen and Kristiansen, 2005; Douay *et al.*, 2008). With the aim of maintaining remediated soils for plant production without great modifications of their physicochemical and biological parameters, chemical remediation can be a promising technology for the immobilisation of metals in kitchen garden soils and may provide a long-term remediation solution if low solubility minerals and/or stable precipitates are produced (US Environmental Protection Agency, 1997; Vangronsveld and Cunningham, 1998). Natural organic amendments like sheep manure, biosolid compost, vegetable waste, waste compost, vermicompost and biosolid compost (McCoy, 2015; Wiszniewska *et al.*, 2016), industrial products like fly ash, lime mud from papermaking process and biochar (Janus *et al.*, 2015; Brännvall and Kumpiene, 2016; Rizman *et al.*, 2016; Zhang *et al.*, 2016) and inorganic compounds like lime, metal oxides and phosphates (Miretzky and Cirelli, 2010; Komárek *et al.*, 2013; Gul *et al.*, 2015; Hafsteinsdóttir *et al.*, 2015; Mahar *et al.*, 2015) have been widely used for the chemical immobilisation of As, Cd, Pb and Zn. In this way, mixture of amendments can be recommended for soils contaminated by metals and metalloids due to the benefits from their synergistic interactions (Ram and Masto, 2014). As a consequence, the choice of the chemical amendment is primordial, and soil reusing has to be taken into account.

Numerous studies in relation with Cd and Pb immobilisation reported the use of chemical amendments like composts, clay minerals, zeolites, ashes, oxides, limestone and phosphates. Among all of these amendments, phosphates have been used in acidic and calcareous soils. It is well known that relationships between phosphates and metals depend on the physicochemical parameters of the treated soils, the speciation of metals and the source of phosphates as well as their solubility. These mechanisms are mainly based on adsorption, surface complexation, precipitation, coprecipitation and/or ion exchange processes (Miretzky and Fernandez-Cirelli, 2008; Mignardi *et al.*, 2013; Koptsik, 2014; Mahar *et al.*, 2015). According to Bolan *et al.* (2014), precipitation as metal-phosphate has been proved one of the main mechanisms for the immobilization of Cd and Pb in soils. The metal-phosphate compounds are very stable and present a low solubility over a wide range of pH, making this technique an attractive method for Cd contaminated soils and particularly for soils contaminated by Pb due to the fact that this metal is known to form pyromorphite, one of the most thermodynamically stable Pb minerals. This probably explains why the

addition of phosphates is one of the best management practices suggested by the US Environmental Protection Agency for Pb-contaminated soils (Chrysochoou *et al.*, 2007). Although using phosphate amendments has benefits, an addition of phosphate at high rates may result in an excess of available P for plant and thus may cause eutrophication of groundwater and surface waters (Melamed *et al.*, 2003; Brown *et al.*, 2004; Park *et al.*, 2011). On the other hand, it was shown that low-molecular-weight organic acids (LMWOA) in combination with biological activities may reduce or inhibit the formation of pyromorphite by precipitation (Debelo *et al.*, 2013). To avoid these phenomena, recent studies showed that it was possible to reduce the environmental availability of metals using two types of P sources, one from soluble phosphates and the other from insoluble phosphates (Basta *et al.*, 2001; Hettiarachchi and Pierzynski, 2002; Cao *et al.*, 2003; Zhu *et al.*, 2004; Waterlot *et al.*, 2011b; Zupančič *et al.*, 2012). The great interest in using this type of phosphate mixture arises from the reduction of the impacts of phosphate compounds on soil pH and the concentrations of soluble and available P, as well as the promotion of pyromorphite formation by dissolution of phosphates followed by precipitation to form new metal-phosphate compounds instead of precipitation alone (McGowen *et al.*, 2001; Cao *et al.*, 2003; Zhu *et al.*, 2004; Chen *et al.*, 2007; Waterlot *et al.*, 2011b; Tang and Yang, 2012; Austruy *et al.*, 2014; Shahid *et al.*, 2014). This is particularly the case in the soil rhizosphere due to the release of LMWOA from roots and microorganisms. The main objective of the present study was to evaluate the effects of an equimolar P mixture of diammonium phosphate (DAP) and hydroxyapatite (HA) on the environmental availability and phytoavailability of Cd and Pb in kitchen garden soils with moderate and high carbonate contents.

## MATERIALS AND METHODS

### *Study site, sampling strategy and soil analysis*

Two kitchen garden soils (T1 and T2) with different physicochemical characteristics were chosen in an area highly contaminated by the past emissions of two Pb and Zn smelters, Metaleurop Nord at Noyelles-Godault and Umicore at Aubry, located in the North of France. An agricultural soil (T3), located far enough from any industrial development, urban area or major road, was selected as the reference soil. Three other kitchen garden soils (T4, T5 and T6) were sampled near a former lead smelter in the region of Île-de-France (Us), and for this region, an uncontaminated kitchen garden

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