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Soil acidification increases metal extractability and bioavailability in old orchard soils of Northeast Jiaodong Peninsula in China

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ABSTRACT

The bioavailability of Cu, Zn, Pb and Cd from field-aged orchard soils in a certified fruit plantation area of the Northeast Jiaodong Peninsula in China was assessed using bioassays with earthworms (*Eisenia fetida*) and chemical assays. Soil acidity increased with increasing fruit cultivation periods with a lowest pH of 4.34. Metals were enriched in topsoils after decades of horticultural cultivation, with highest concentrations of Cu (132 kg^{-1}) and Zn (168 mg kg^{-1}) in old apple orchards and Pb (73 mg kg^{-1}) and Cd (0.57 mg kg^{-1}) in vineyard soil. Earthworm tissue concentrations of Cu and Pb significantly correlated with 0.01 M $CaCl_2$-extractable soil concentrations ($R^2 = 0.70$, $p < 0.001$ for Cu; $R^2 = 0.58$, $p < 0.01$ for Pb). Because of the increased bioavailability, regular monitoring of soil conditions in old orchards and vineyards is recommended, and soil metal guidelines need reevaluation to afford appropriate environmental protection under acidifying conditions.

1. Introduction

Horticultural soils may contain elevated metal concentrations as a result of the application of agrichemicals and soil amendments. Soil acidification may increase the bioavailability of these metals worsening its contamination condition. Soil Cu accumulation has been reported in the surface layer of European and Australian vineyard soils (Delusia et al., 1996; Chopin et al., 2008; Wightwick et al., 2008). A similar increase in Cu has occurred in citrus orchards and avocado orchard soils with an established history of copper-based fungicide use, with current concentrations ranging between 110 and 1500 mg kg^{-1} Cu (Fan et al., 2011; Merrington et al., 2002). Several studies in other countries have shown that substantial quantities of Pb and As may accumulate in orchard topsoils as a result of repeated lead arsenate application (Pendergrass and Butcher, 2006; Udovic and McBride, 2012; Hood, 2006).

The region of the Northeast of Jiaodong Peninsula in China, which can be considered part of the so called “fruit belt” of North Latitude 36 of the world, has a long tradition of intensive horticultural crop production. In this region, some old orchards and vineyards are being planted with other crops for animal and human consumption. Subsequent changes in land use can promote a potential problem of metal bioavailability and toxicity that is strongly influenced by soil properties. Acidification was severe in orchard soils of the Northeast of Jiaodong Peninsula according to a preliminary investigation in 2007–2009, which indicated that topsoil pH in 60.4% of the 268 investigated sites was less than 5.5 (with 27.2% less than 4.5). Nitrogen fertilization and irrigation are crucial for sustainable fruit production. Excessive application of N may, however, produce more acidity than actually necessary. Fruit trees also take up a large amount of cations from the soils each growing season (Tang et al., 2000). Over time, soils may become more acid due to the removal (loss) and leaching of exchangeable bases such as calcium and magnesium, which is especially pronounced in the coastal sandy textured soils with low nutrient-holding buffering capacity (i.e., the Northeast of Jiaodong Peninsula). Surface soil acidification and the effect on metal bioavailability have been investigated in laboratory and field studies (Allen, 2002;

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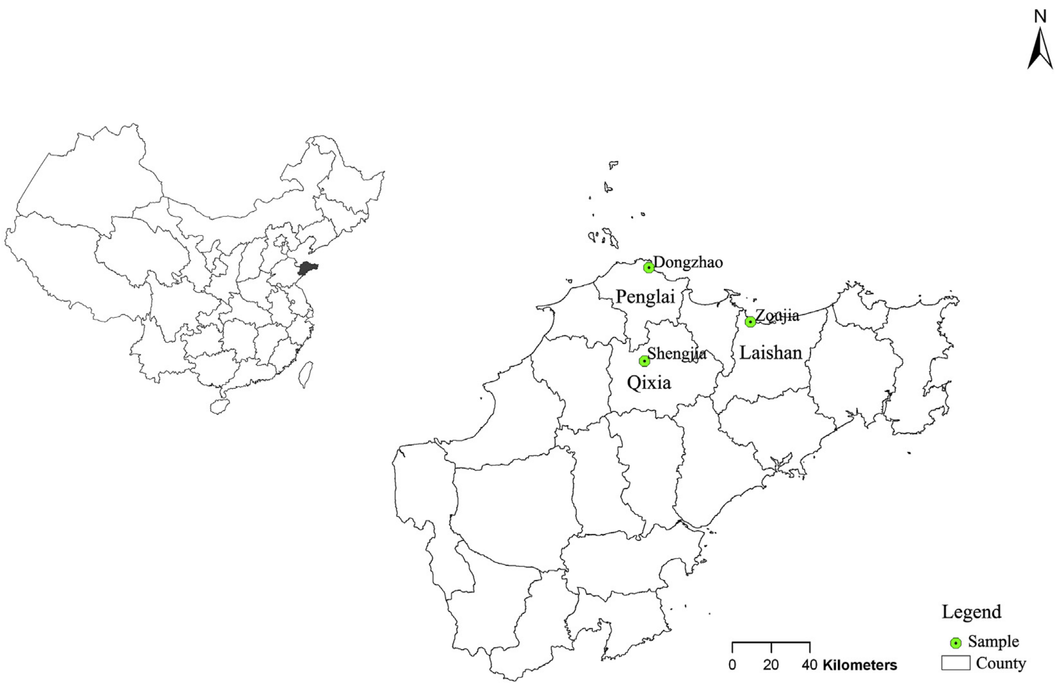

Fig. 1. Location of the study area and the soil (Udic Luvisols) sampling sites of Qixia, Penglai and Laishan in Northeast of Jiaodong Peninsula of China. The region has a long tradition of intensive horticultural crop production.

Peijnenburg et al., 1999; Guo et al., 2011). However, there is little knowledge of subsoil acidity in the Northeast of Jiaodong Peninsula fruit-growing area, although topsoil acidification has become a major problem in soils of intensive Chinese agricultural systems (Guo et al., 2010). As the root system of most species of tree fruit are in the subsurface soil horizon, it is most vital to assess soil acidification in this layer and address its effects on metal bioavailability.

In general, there are two empirical approaches to assess availability of contaminants: chemical and biological testing. Of all chemical measures of bioavailability, chemical extraction techniques are commonly used as has been reviewed by Peijnenburg et al. (2007). Measuring body residues in soil organisms provides a better indication of metal bioavailability than using total metal concentrations in soil (Li et al., 2009; Conder and Lanno, 2003; Ma, 2005). Earthworms, because of their close contact with soil, are suitable organisms for assessing the bioavailability of contaminants in soil and the potential for these contaminants to enter terrestrial food chains (Spurgeon and Hopkin, 1999; Lock and Janssen, 2001; Vijver et al., 2006; van Gestel, 2012; OECD, 2004).

In this study, field and laboratory investigations were conducted to (1) study the accumulation and distribution of selected metals (Cu, Zn, Pb and Cd) in the surface and subsurface soils of the fruit production region of Northeast of Jiaodong Peninsula in China; (2) determine whether intensive horticulture cultivation (including multiple management practices like tillage, fertilization, and irrigation) affects orchard and vineyard soil pH in the surface and subsurface layers; (3) evaluate the importance of soil solution pH on metal distribution and chemical speciation; and (4) estimate the availability of metals in soils using various chemical extractions as well as earthworm bioassays.

2. Materials and methods

2.1. Study area

The region of the Northeast of Jiaodong Peninsula has a marine climate, with humid air, ample sunlight and a high annual rainfall of 650–850 mm, and is propitious to the growing of tree fruits, such as apple and grapevine. The soils in this area are brown soils (Udic Luvisols). The area has a long tradition of intensive horticultural crop production and has become a main region for apple production in China and one of the main regions for grape growing throughout the world.

2.2. Soil sampling

The soils were sampled in 2011 using a 0.10-m-diameter drill to collect samples from the vineyards in Penglai and two types of orchards, apple in Qixia and sweet cherry in Laishan. The studied orchards and vineyards were classified into three main categories based on their cultivation age, namely: Young, Adult and Old. Young orchards and vineyards were defined as those planted less than 10 years, Adult planted 10–30 years and Old planted more than 30 years ago. For each of the three cultivation age groups, triplicate soil samples from different locations were randomly collected at depths of 0–20 cm and 20–40 cm, representing surface and subsurface horizons. To ensure representativeness in the sample from each triplicate, five soil sub-samples were randomly collected in the middle of rows located in the central portion of the stand. In this way, 15 soil sub-samples were obtained from each group of vineyards and orchards and subsequently mixed into a composite sample to obtain an accurate representation of the plot.

Samples were air-dried, ground and passed through a 2 mm Nylon sieve prior to analyses. Soil sample sites are shown in Fig. 1 in relation to the spatial distribution of vineyard and orchard sites.

2.3. Physicochemical characterization of the soils

The pH of soil samples was determined in triplicate at a 1:2.5 soil to water (w/v) ratio after shaking for one hour, using a glass electrode (Metrohm780 pH meter, Herisau, Switzerland). Total soil organic C was determined by combustion using a C/N analyzer (Vario MAX CN Macro Elemental Analyzer; Elementar Analysen System GmbH, Germany). A laser analyzer Mastersizer 2000 (Malvern Instruments) with Hydro MU adapter was used to determine the particle-size distribution of soil samples.

2.4. Soil metal analysis and extraction

Soil samples were digested in a mixture of concentrated HNO_3–HF–$HClO_4$ (v/v/v = 5:3:2; ultra pure grade, Sigma–Aldrich Shanghai Trading Co., Ltd, Shanghai, China), using a microwave digestion system (MAR-5, CEM Corporation, Matthews, NC, USA). Metal concentration was then determined by inductively coupled plasma-mass spectrometry (ICP-MS; Agilent 7500i, Agilent Technologies Co. Ltd, USA). A certified reference soil (GBW07306, Polluted soil, State Bureau of Technical Supervision, People's Republic of China) was included as a means of quality control of the analysis. For all metals (Cd, Cu, Pb, and Zn) analyzed, measured concentrations were within 10% of the reported certified concentrations.