



Insights into the chemical partitioning of trace metals in roadside and off-road agricultural soils along two major highways in Attica's region, Greece



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ABSTRACT

We report in this study the magnetic properties and partitioning patterns of selected trace metals (Pb, Zn, Cu, Cd, Ni) in roadside and off-road (> 200 m distance from the road edge) agricultural soils collected along two major highways in Greece. Sequential extractions revealed that the examined trace metals for the entire data set were predominantly found in the residual fraction, averaging 37% for Cd up to 80% for Cu. Due to the strong influence of lithogenic factors, trace metal pseudototal contents of the roadside soils did not differ significantly to those of the off-road soils. Magnetic susceptibility and frequency dependent magnetic susceptibility determinations showed a magnetic enhancement of soils; however, it was primarily related to geogenic factors and not to traffic-derived magnetic particles. These results highlight that in areas characterized by strong geogenic backgrounds, neither pseudototal trace metal contents nor magnetic properties determinations effectively capture traffic-related contamination of topsoils. The vehicular emission signal was traced by the increased acid-soluble and reducible trace metal contents of the roadside soils compared to their off-road counterparts. In the case of Cu and Zn, changes in the partitioning patterns were also observed between the roadside and off-road soils. Environmental risks associated with agricultural lands extending at the margins of the studied highways may arise from the elevated Ni contents (both pseudototal and potentially mobile), and future studies should investigate Ni levels in the edible parts of plants grown on these agricultural soils.

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1. Introduction

Roadside soils are enriched in several classes of organic and inorganic contaminants, including trace metals (Sutherland and Tolosa, 2000). The sources of trace metals in these soils are related to road surfaces, vehicular traffic, as well as road traffic control and maintenance operations. In detail, these include: road surface wear, road paint degradation, vehicle wear of tires, body and brake linings, lubricating oils, corrosion of galvanized surfaces of crash barriers and road signs, as well as particulate emissions (Sutherland and Tolosa, 2000; Thorpe and Harrison, 2008; Werkenthin et al., 2014). Trace metals could be easily transported to the roadside environment through re-suspension and aeolian transport, surface runoff and spray water, and road drainage systems, leading to adverse effects on terrestrial and aquatic systems. There

is a general concession that these metals decrease in concentration with distance from the roadway (Werkenthin et al., 2014). The chemical form in which the metals in roadside soils occur, however, has not been investigated so far and consequently there is a gap of knowledge for their mobilization.

Large scale urbanization and outward growth of cities has resulted in the expansion of transport systems, assuring the continued need for transportation of people, natural resources and products. The heavy traffic and the fast movement of vehicles can produce a large amount of road-deposited particles contaminated with trace metals that are eventually incorporated in the roadside soil (Christoforidis and Stamatis, 2009; Huber et al., 2016; Nazzari et al., 2013). Agricultural lands extending at the margins of highways are not uncommon in modern sprawling cities (e.g. Morse et al., 2016; Ward, 1990). Trace metal contamination of agricultural soils is of great environmental concern because of the possible long-term phytotoxic effects and food chain contamination. Therefore, assessing the availability of trace metals relevant to plant uptake is essential for evaluating agricultural implications

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and managing human health risks (Chen et al., 2016; Kelepertzis et al., 2015). Furthermore, health risks may arise to urban and suburban populations through respiration and inadvertent ingestion of contaminated dusts re-suspended by wind, or, street sweeping operations. The degradation of groundwater quality is another issue of concern when road runoff water percolates through the roadside soils, resulting to the dispersion and mobilization of contaminated soil particles (Huber et al., 2016; Werkenhain et al., 2014).

The potential mobility and ecotoxicity of trace metals are determined by their specific binding form and coupled reactivity, rather than by their total concentrations (Peijnenburg et al., 2007). This is particularly valid for anthropogenic metals, because large amounts of them are associated with less stable geochemical phases, with severe consequences regarding their availability to plant uptake and migration into groundwater aquifers. Furthermore, metal release-retention processes are largely controlled by the soil properties. Changes of the physical biogeochemical soil conditions, including leaching and ion exchange processes, changes of soil pH and redox potential, soil organic matter decomposition and microbial activity may have a profound effect on trace metals mobility, thus their bioavailability.

Sequential extractions, despite their limitations and experimental uncertainties reviewed by many authors (e.g. Bacon and Davidson, 2008; Tack and Verloo, 1995), are considered a powerful tool in assessing the strength of trace metals association with the solid phase and predicting their behavior and fate under changing environmental conditions. These methods are based on the rational use of a series of successive chemical treatments, each being more drastic, or of different nature from the other (Gleyzes et al., 2002). In general, the strength of the treatment increases through the steps of the procedure. Hence, the trace metals extracted early in the procedure are those most weakly bound to the solid phase, consequently, those with the greatest potential of mobility than those released later (Bacon and Davidson, 2008). Extraction steps also intend to simulate the modifications of environmental conditions that could affect metal-binding in the solid surfaces, such as acidification, reduction and oxidation (Bacon and Davidson, 2008; Gleyzes et al., 2002). Sequential extraction techniques have been applied in various environmental sampling media, such as in agricultural soil (examples given by Abollino et al., 2002; Sungur et al., 2015; Yang et al., 2013), mining soils (e.g. Li and Thornton, 2001), as well as roadside and industrially contaminated street dusts (Yildirim and Tokalioglu, 2016).

Magnetic techniques have been successfully applied in urban environments to detect and spatially outline automobile-related, trace metal contamination of soils (e.g. Golden et al., 2015; Lu et al., 2011; Yang et al., 2010). The relationship between magnetic signals and trace metals contents is governed by two major principles: i) magnetic minerals and trace metals derive from a common process (e.g. combustion), and ii) magnetic minerals serve as the substrates for trace metals (deriving probably from a different source), through adsorption, complexation, and co-precipitation processes (Lu et al., 2011; Scoullou et al., 2014). The sources of magnetic minerals in road surfaces were investigated by Beckwith et al. (1986) and included corrosion of surface constructional materials, exhaust particulates, and corroded bodywork, with vehicular rust particulates (wheel hub particulates and corroded paintwork) contributing significantly to the overall magnetic signal.

Although traffic-related trace metal contamination of roadside soils is recognized as a major environmental problem and several studies have been conducted along European main roads and highways (e.g. Kluge and Wessolek, 2012; Pagotto et al., 2001; Ward et al., 1977; Ward, 1990), there are only few studies of roadside soils that included sequential extraction determinations

(Bäckström et al., 2004; Harrison et al., 1981; Norrström and Jacks, 1998). More importantly, peer-reviewed research on the speciation of trace metals associated to farming on marginal land adjacent to highways is limited (Bacon and Hewitt, 2005). From a human health risk perspective, a question is if there are differences in the chemical partitioning of metals between agricultural soils near to major highways, and agricultural soils away from the highways. The present study, by combining chemical analyses and magnetic techniques, was initiated to examine traffic-related pollution at the marginal agricultural soils along two major highways of the Metropolitan city of Athens in Greece. The specific objectives of the study are: (a) to investigate the potential of magnetic susceptibility measurements for detecting the roadside trace metal contamination in the studied agricultural soils, (b) to compare the levels and partitioning of trace metals into the major fractions of soil components at distances 0–10 m and > 200 m off the highways, c) to evaluate the potential risks arising from the agricultural activities of the roadside soils.

2. Materials and methods

2.1. Sampling and samples pre-treatment

Samples were collected from roadside and off-road agricultural soils located in ten sites in the vicinity of two major highways in Greece (Fig. 1). Five sites were selected from the National Road Athens-Lamia, which is the oldest and most important highway in Greece (this highway connects the country's largest cities, Athens and Thessaloniki). Another five sites were selected from the Attiki Odos highway, which forms part of the urban highway network of Athens metropolitan area, leading to the Athens International Airport. In both highways, the majority of cultivated plants were primarily vines, followed by olive trees, clover, vegetables and walnut trees. The Attiki Odos along with the Athens International Airport to which it connects opened in 2001. Anthropogenic enrichment for Pb (median value of 67 mg/kg) in agricultural soils surrounding the Athens International Airport was identified in the study of Kaitantzian et al. (2013). In each site, two soil samples were collected, the first at the closest vicinity to the road, at distances ranging from 2 to 10 m, (denoted as sample A), and the second one at a distance of approximately 200–400 m from point A at the perpendicular direction, which is denoted as sample B.

Soil sampling was conducted by using a plastic spade at a depth of 0–10 cm. At each site, three sub-samples were collected within an area of 4 m² and mixed to obtain a single composite sample of 1–1.5 kg. A total of 20 samples were collected and stored in plastic bags for transportation to the laboratory. Thereafter, the soil samples were air-dried 3 days and then gently disaggregated and sieved through a 2 mm sieve in order to remove granules, plant debris and litter. These sieved soil samples were used for the determination of soil pH, electrical conductivity and magnetic susceptibility, as well as texture analyses. All other analyses were conducted in representative sub-samples that were finely ground in a mortar and sieved to pass through a 0.5 mm sieve.

2.2. Reagents and apparatus

Reagents of analytical purity (Merck, Germany) and Deionized Distilled Water (DDW; 18.2 MΩ/cm resistivity) were used in the analytical procedures. Plastic and glassware were soaked in 1:1 (v/v) HNO₃: DDW for 24 h, then rinsed with 1% (v/v) HNO₃ and dried in an oven at 40 °C before use. Precautions were taken during all stages of analytical procedures in order to avoid contamination.

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