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# Heavy metals in agricultural soils of the European Union with implications for food safety

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ABSTRACT

Soil plays a central role in food safety as it determines the possible composition of food and feed at the root of the food chain. However, the quality of soil resources as defined by their potential impact on human health by propagation of harmful elements through the food chain has been poorly studied in Europe due to the lack of data of adequate detail and reliability. The European Union's first harmonized topsoil sampling and coherent analytical procedure produced trace element measurements from approximately 22,000 locations. This unique collection of information enables a reliable overview of the concentration of heavy metals, also referred to as metal(loid)s including As, Cd, Cr, Cu, Hg, Pb, Zn, Sb. Co, and Ni. In this article we propose that in some cases (e.g. Hg and Cd) the high concentrations of soil heavy metal attributed to human activity can be detected at a regional level. While the immense majority of European agricultural land can be considered adequately safe for food production, an estimated 6.24% or 137,000 km$^2$ needs local assessment and eventual remediation action.


## 1. Introduction

The heavy metal (HM, also referred to in scientific literature as metal[oid]) contamination of soil is one of the most pressing concerns in the debate about food security and food safety in Europe (CEC, 2006a) and globally (Kong, 2014). A recent review by Peralta-Videa et al. (2009) summarizes the impact of heavy metal from food origin on human health as well as the mechanism of uptake, transformation and bioaccumulation of heavy metals by plants.

The number of contaminated sites in the European Union (van Liedekerke et al., 2014) and the area affected by different kinds of pollution, of which the remediation would cost €17.3 billion annually (CEC, 2006b) underlines the extent of the problem in the continent. Apart from soil contamination which may lead to the degradation of water quality and a series of negative impacts on the environment (Mulligan et al., 2001; Rattan et al., 2005), the propagation of heavy metals throughout the food chain have serious consequences for human health (Järup, 2003). Despite of the importance of HM contamination, so far there has been no sufficient data to provide a reliable view on the real extent of the problem in Europe and worldwide. FOREGS data produced by the EuroGeoSurvey (Salminen, 2005) and the derived continuous map sheet (Lado et al., 2008) have been the most comprehensive source of information to date. However, the low sampling density (1 site/5000 km$^2$) of the FOREGS study (Demetriades et al., 2010) allows only limited interpretation apart from the provision of a continental-scale overview without the possibility of comparing the concentrations by land use type.

The LUCAS Topsoil Survey, with its 1 site/200 km$^2$ sampling density opened new prospect in this regard. The survey represents the first effort to build a consistent spatial database of soil properties for environmental assessments ranging from regional to continental scale on all major land use types across Europe (Tóth et al., 2013). As the inputs of HM to soils are accumulated in the topsoil (Hou et al., 2014) and crop and meadow grass nutrient uptake also takes place predominantly from this zone (Kismányoky and Tóth, 2010), the LUCAS Topsoil Survey presents an adequate information base to assess the HM load to the environment and its potentials to enter the food chain. The standard sampling and analytical procedures of the Survey – with the analysis of all soil samples being carried out in a single laboratory – provides a basis for an EU wide harmonized soil monitoring scheme as well.

In this paper a detailed analysis of the HM content in agricultural topsoils of the European Union is delivered. The analysis covers the main potentially toxic elements, namely As, Cd, Cr, Cu, Hg, Pb, Zn, Sb, Co and Ni. Soil heavy metal content was assessed against element-specific thresholds of contamination and remediation needs. While delivering a new insight into the level of soil HM contamination and highlighting the needs to intensify monitoring or taking remediation actions to eliminate risks to human health in specific regions, the study does not cover aspects like the bioavailability of elements by various

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plant species or the potential differentiated impact of elemental speciation to ecological conditions or human health.

## 2. Materials and methods

### 2.1. Soil sampling

With the scope of creating the first harmonized and comparable data on soil at European level to support policymaking Eurostat together with the European Commission's Directorates-General for Environment (DG ENV) and the Joint Research Centre (JRC) designed a topsoil assessment component ('LUCAS-Topsoil') within the 2009 and 2012 LUCAS surveys (Tóth et al., 2013; Tóth et al., 2015). The LUCAS Programme itself assesses the land use and land cover parameters that are deemed relevant for agricultural policy. Since 2006 the sampling design is based on the intersection of a regular grid covering the territory of the EU (Eurostat, 2015a). Around 220,000 points are periodically visited as control points for the survey. The LUCAS 2009 and 2012 surveys included topsoil sampling at around 10% of those points, which were visited for land use and land cover assessment in 27 EU Member States (all current EU countries excluding Croatia, which joined the EU in 2014). As a result, topsoil samples were collected from some 22,000 points using a standardized sampling procedure. In order to secure the most reliable overview of soil properties in European regions, a multi-stage stratified random sampling approach (McKenzie et al., 2008) was chosen. Altitude, slope, aspect (orientation of the slope), slope curvature and land use were considered for the stratification of the survey points. It is worth noting that the geographical coordinates of some samples (<5% of the collection) were not fully recorded, or the records had low reliability. These samples were not considered in our analysis. Regions with inadequate sample size (less than 5 samples from agricultural land) were omitted from the current study as well.

Samples were collected from the designated locations by a process of composite sampling. Five soil subsamples were taken and mixed together at each sampling. These composite soil samples, weighting about 0.5 kg each, were dispatched to a central laboratory for physical and chemical analyses.

### 2.2. Methods of laboratory analysis

The laboratory analysis of the soil samples for the basic soil parameters followed standard procedures (Tóth et al., 2013). After the analysis of the basic soil parameters – which project concluded in 2012 – soil tests for heavy metal content, including As, Cd, Co, Cr, Cu, Ni, Pb, Sb and Zn were carried out. Elements were analyzed by inductively coupled plasma–optical emission spectrometry. Two certified reference materials (BCR 141R, Calcareous Loam Soil, and NIST 2711, Montana Soil) were used to compare the accuracies of the two digestion procedures. In the first phase of the HM analysis comparative tests were performed using two digestion methods on a subset of 500 samples (Comero et al., 2015). The standard method (ISO, 1995) using aqua regia as an extracting agent was matched with one using microwave-assisted acid digestion (ECS, 2010) and the same detection methods, employing ICP–OES (inductively coupled plasma optical emission spectrometer) for the above listed elements. Based on the reliable correspondence between the measured concentrations by the two methods and considering the advantages of the microwave assisted approach (Comero et al., 2015), all samples were analyzed using the prEN16174 (ECS, 2010) procedure. The unit of measurement was mg/kg for As, Cd, Cr, Cu, Pb, Zn, Sb, Co and Ni, with detection limits 2.84, 0.07, 0.32, 0.26, 1.16, 2.12, 0.81, 0.15 and 0.27 mg/kg respectively.

As a result of the analytical procedure we obtained the concentrations of the studied elements. These are expressed by their elemental weight in milligram per 1 kg of soil. No elemental speciation was measured.

In order to enable a full spatial analysis of the results, samples with concentrations below the detection limit were characterized with a value equal to the half of the detection limit. Although this approach might be misleading when mapping the presence of the elements in soil and might cause bias in other applications as well, it seemed to be a sufficient solution for our purposes. The fact that the detection limits are an order of magnitude smaller concentrations than what is considered to have any ecological or health risk (Table 1) confirms the adequacy of the approach.

### 2.3. Assessment of soil heavy metal contamination and remediation needs

European countries have a number of approaches to define risk levels associated with different concentrations of heavy metal in soil (Carlon et al., 2007; Ferguson, 1999). After investigating the options presented by the various approaches and thresholds applied by them, we chose the standards set in the Finnish legislation for contaminated soil (Ministry of the Environment — MEF, Finland, 2007). The Finnish standard values represent a good approximation of the mean values of different national systems in Europe (Carlon et al., 2007) and India (Awasthi, 2000) and they have been applied in an international context for agricultural soils as well (UNEP, 2013). The Finnish legislation sets concentration levels by each hazardous elements to identify soil contamination and remediation needs. It sets lower and higher concentration levels indicating the need for different actions if exceeded. Higher concentration levels are defined by major land uses, i.e. for industrial or transport sites and for other land uses. The so called "threshold value" is equally applicable for all sites and it indicates the need for further assessment of the area. In areas where background concentration is higher than the threshold value, background concentration is regarded as the assessment threshold. The second concentration level is the so-called "guideline value". If this is exceeded, the area has a contamination level which presents ecological or health risks. Different guideline values are set for industrial and transport areas (higher guideline value) and for all other land uses (lower guideline value). With the aim to characterize the soil contamination statuses of European soils, we classified the LUCAS topsoil samples by their heavy metal concentration values by elements using the threshold value and guideline value standards of the Ministry of Environment of Finland (2007) into four categories. Soil samples in the first category have no detectable content or the concentration is below the threshold value set by the MEF. The concentration of the investigated element in the second category is above the threshold value, but below the lower guideline value. The third category includes samples in which the concentration of one or more element exceeds the lower guideline value but is below the higher guideline value while the fourth category includes samples having concentrations above the higher guideline value. For assessing agricultural land we applied the threshold and lower guideline values for samples

**Table 1**
Threshold and guideline values for metals in soils (extract; MEF, 2007).

| Substance (symbol) | Threshold value mg/kg | Lower guideline value mg/kg | Higher guideline value mg/kg |
|---|---|---|---|
| Antimony (Sb) (p) | 2 | 10 (t) | 50 (e) |
| Arsenic (As) (p) | 5 | 50 (e) | 100 (e) |
| Mercury (Hg) | 0.5 | 2 (e) | 5 (e) |
| Cadmium (Cd) | 1 | 10 (e) | 20 (e) |
| Cobalt (Co) (p) | 20 | 100 (e) | 250 (e) |
| Chrome (Cr) | 100 | 200 (e) | 300 (e) |
| Copper (Cu) | 100 | 150 (e) | 200 (e) |
| Lead (Pb) | 60 | 200 (t) | 750 (e) |
| Nickel (Ni) | 50 | 100 (e) | 150 (e) |
| Zinc (Zn) | 200 | 250 (e) | 400 (e) |
| Vanadium (V) | 100 | 150 (e) | 250 (e) |

The guideline values have been defined on the basis of either ecological risks (e) or health risks (t). If the risk of groundwater contamination is higher than normal in concentrations below the lower guideline value, the substances are marked with the letter p.