



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 km² needs local assessment and eventual remediation action.

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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 km²) 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 km² 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.

originating from agricultural areas and the threshold values and higher guideline values for all samples.

2.4. Comparison of HM concentrations across regions and land use types

The LUCAS topsoil database provides a range of opportunities to compare HM concentrations across land use types and management practices, countries, regions, climatic and geological factors and other variables, including socioeconomic data. The primary aim of our current study was to perform a reconnaissance in the soils of the European Union in general and in agricultural soils in particular. Therefore we analyzed agricultural land use types in comparison with all land uses with regards to HM concentrations.

The LUCAS dataset was subsampled to extract samples from agricultural land use types, namely cropland and grassland. All remaining samples were regarded as from “other land uses”. A series of descriptive statistics and multiple comparison tests were performed to assess the topsoil data from agricultural land use and other land uses in different climatic regions and countries of the EU. One-way ANOVA tests were also performed in specific cases to assess if there were significant differences between larger geographical regions (i.e. Eastern and Western Europe) or land use types concerning their soil HM concentration, on a 0.05 confidence level.

For the regional analysis in the EU the so-called basic regions for the application of regional policies (NUTS2; Eurostat, 2015b) were used. The spatial dataset of the NUTS2 units was accessed from the Eurostat website. As the area of the NUTS2 regions in Europe differ greatly and not all statistical regions had sufficient number of samples to draw reliable conclusions from, we analyzed only those regions from which at least five soil samples were taken in the LUCAS survey. Heavy metals in the topsoil of 248 regions were studied.

3. Results and discussion

3.1. Overview of heavy metals' concentrations in the soils of the European Union

The soil heavy metal assessment in the European Union shows a quite diverse pattern both for geographic variability and the distribution of samples by the different concentrations of various heavy metals (Fig. 1).

Results of the analyses of all heavy metals for each soil sample were combined to see, which samples have concentrations above threshold or guideline values of any one or more elements. Figs. 2–5 display the percentage of samples having high concentration of any heavy metals, by NUTS2 regions of the EU. Most regions in the EU have very high percentages of samples which have concentrations above the investigation thresholds, both on their entire land area (Fig. 2) and on their agricultural land (Fig. 3). Regional differences can be observed in the continental overview. North-eastern Europe and Eastern-Central Europe is less affected by high concentrations of heavy metals, while most soil samples in Western-Europe and the Mediterranean have concentration above the investigation threshold of least one kind of heavy metal. This alarming figure urges for the establishment of detailed monitoring of soil throughout the EU.

Summary statistics (Table 2) also show that agricultural land of the EU has higher percentage of samples with concentration above the threshold value, than other land uses. This figure probably reflects the fact that forest land, which provides the second most LUCAS samples after agricultural areas, are less affected by heavy metal contamination.

The relatively high percentage (6.24%) of samples with any kinds of heavy metal concentration above the guideline value set for agricultural land suggest that an estimated 137,000 km² of agricultural land is affected to a certain degree (Fig. 4). Furthermore, 2.56% of the samples

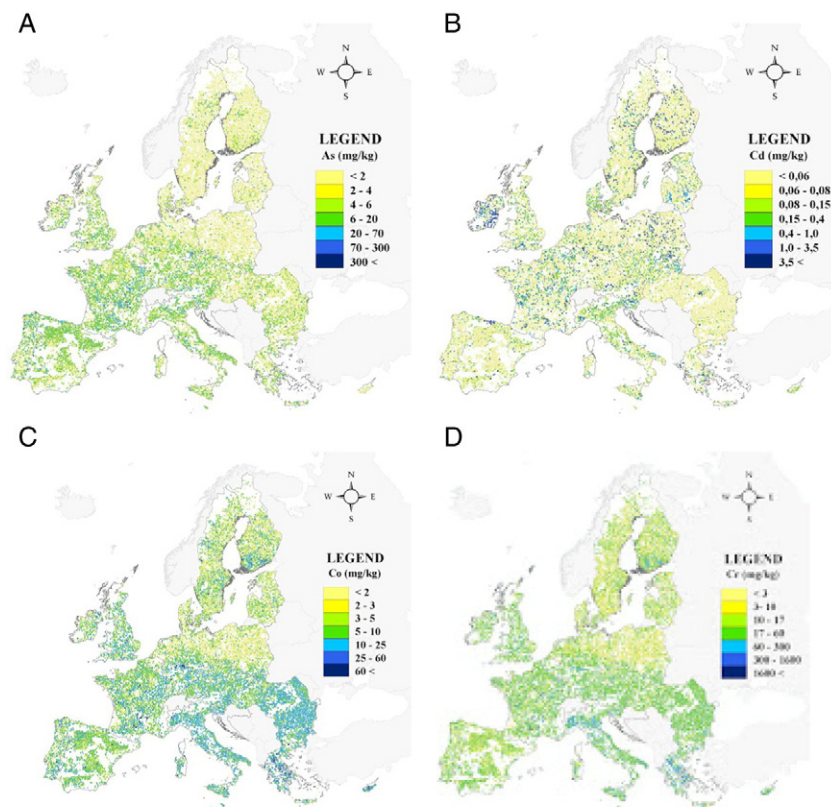


Fig. 1. Spatial representation of heavy metal levels in topsoil of the European Union.

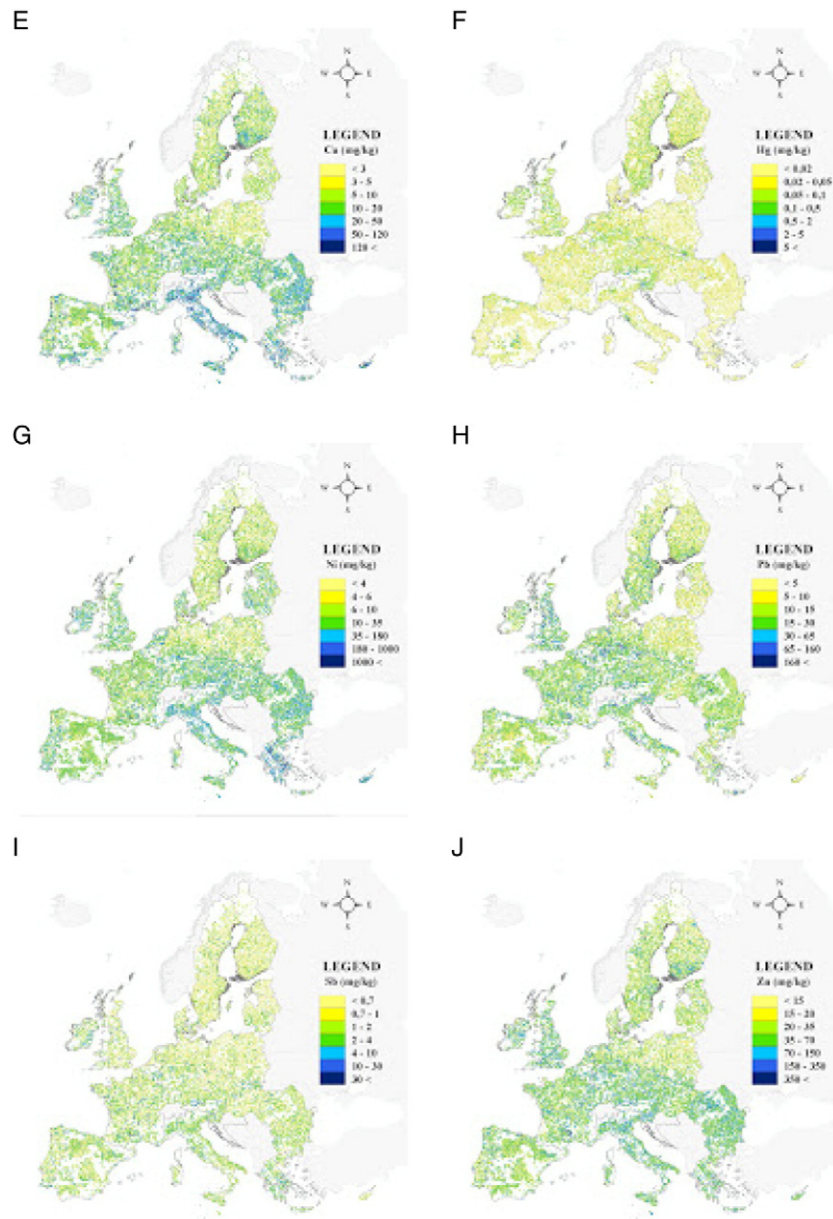


Fig. 1 (continued).

from agricultural land contained heavy metal in concentration which would require remediation also if these were from industrial or transport areas (Fig. 5), based on the applied guideline values.

3.2. Arsenic in topsoils of the European Union

Arsenic in soil is generally considered to be mainly of geological origin, with higher background concentration in clayey soils. However anthropogenic arsenic pollution is quite widespread, as release of arsenic from anthropogenic sources far exceeds those of natural origin (ATSDR, 1999). A previous study by Ursitti et al. (2004) revealed that arsenic does not migrate laterally and its vertical movement is also limited in soils. Our results confirm the dominance of geological reasons behind arsenic concentrations in topsoil on a continental scale, as the main border line between regions with high and low concentration coincides with that of the last glaciation (Fig. S1A, B). Areas of quaternary origin in the north show significantly lower concentrations than most of other regions in Europe. Our findings also suggest that the geomorphology-based explanation of topsoil arsenic is less relevant. A

detailed analysis of samples from the north European region with recent soils developed after the last glaciation showed no significant influence of soil texture on As concentration, either. However, south-eastern Europe, including Hungary, Romania and to some extent Slovakia, Bulgaria and Greece have generally lower levels of arsenic in their topsoil. More than half of the EU statistical regions have samples with As concentration above the investigation threshold concentrations in the majority of the soil samples. With regards to agricultural land, 15% of the regions had more than 1% of their samples with As concentration above the lower guideline value (Table 1), in 7 regions the number of such samples was above 5% and in 3 regions it reached or exceeded 10% (Fig. S1C, D). Only two regions had more than 10% of their samples above the lower guideline value, but these regions had few sampling points, among which 1 and 2 samples were found to be affected. Similar figures were obtained for samples from all major land uses with regards to higher guideline values (Fig. S1E). Furthermore, some agricultural areas, mainly in the Mediterranean countries, have higher As content than allowed by the higher guideline value (Fig. S1F). This fact urges for thorough investigation of the arsenic problem in particular in

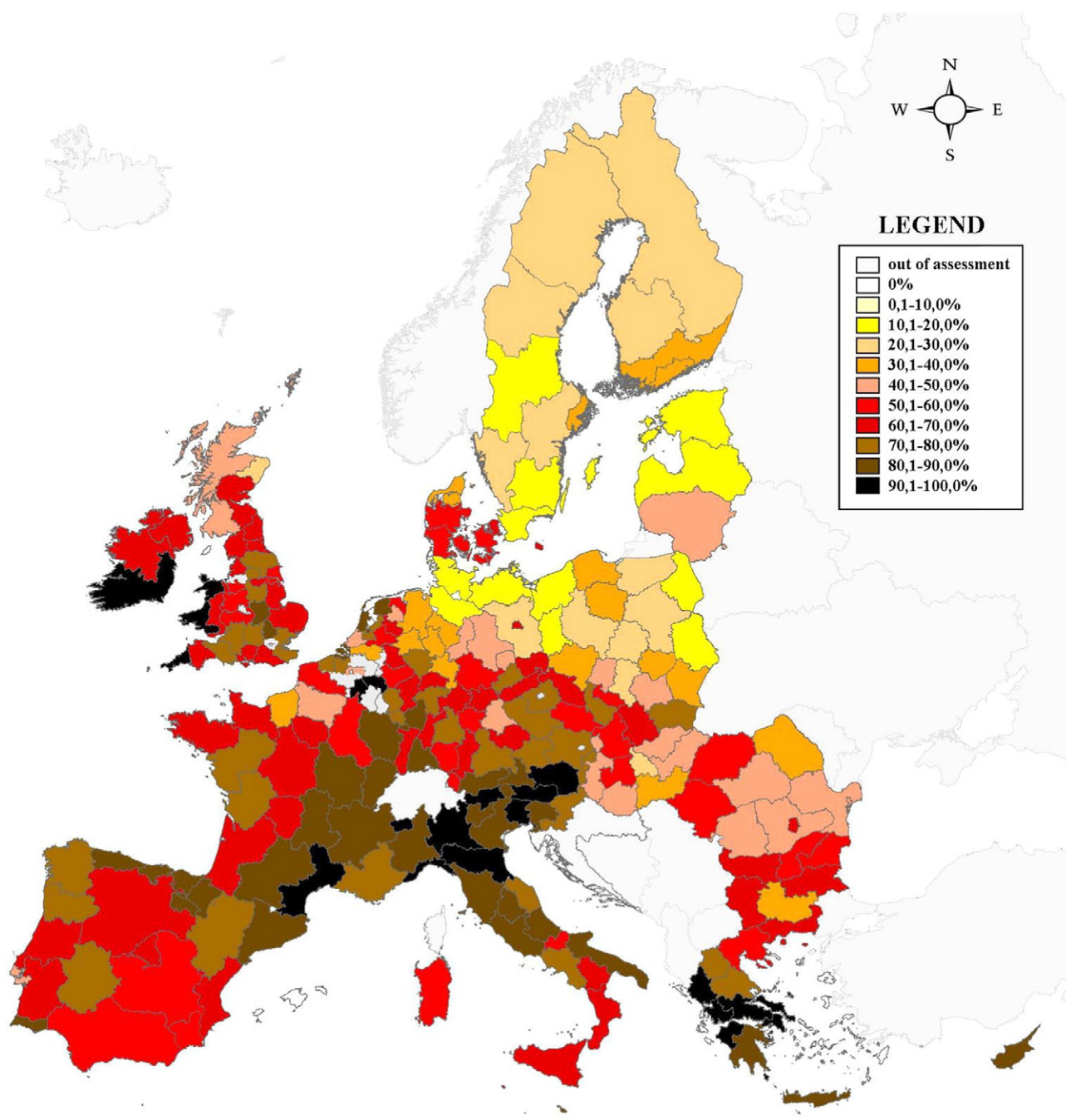


Fig. 2. Percentage of samples with concentrations above the threshold value in all the LUCAS samples.

France, Italy and Spain. The sporadic distribution of samples with high concentration indicates that Arsenic pollution can be a continent-wide problem but only on a small percentage (0.8%) of agricultural areas and in other land uses.

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3.3. Cadmium concentrations in topsoils of the European Union

Cadmium concentrations show a diverse pattern in soils across the European Union (Fig. S2B). Most samples (72.6%) of Europe did not display detectable concentrations of Cd and only 5.5% of the samples have concentrations above the threshold value. However, with the exception

of Estonia and Hungary, whose samples did not display any detectable cadmium contamination, soil samples with Cd concentration above the investigation threshold were found throughout the EU (Fig. S2A). Regions with some of the highest mean cadmium concentration can be found in Ireland and Greece. Nevertheless, we can declare that agricultural areas in Europe are safe from cadmium contamination at the present (Fig. S2D, F). Only isolated cases in France and Spain provided soil samples with concentrations above the guideline values set for land for food production. In some regions the generally higher concentrations – which are still below food safety considerations – might correspond to natural Cd concentrations. However, the LUCAS data call for strict measures to prevent further increase of Cd in soil in many European regions. According to EFSA (2012a) the European population has an average daily Cd intake of 35% the recommended maximum, which intake can be up to 135–208% in some groups of the population.

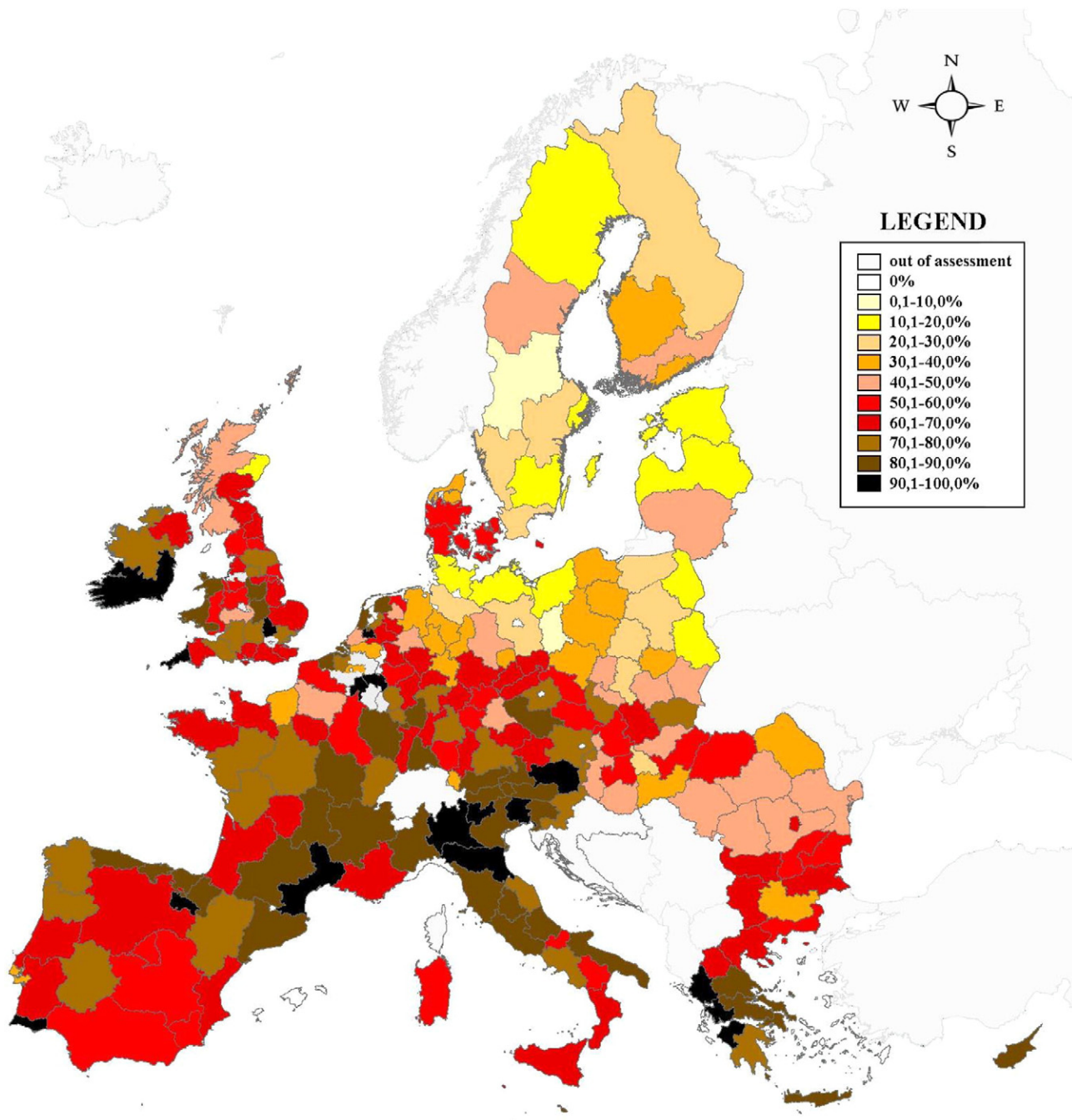


Fig. 3. Percentage of samples with concentrations above the threshold value in LUCAS samples from agricultural land.

As most of this Cd enters the human body through the food materials, which accumulate Cd from the soil, soil protection measures are needed to maintain or improve the current situation by preventing any further Cd contamination e.g. by controlling Cd in phosphorus fertilizers.

Detailed statistical analysis also revealed the significant difference between Cd concentration in agricultural land of Eastern and Western Europe. Data shows higher concentrations of Cd in the agricultural soils of Western Europe (EU15) compared to those in the new member states (EU12). There was no such difference in the data when also assessed against the concentrations of LUCAS samples from forest areas, which suggests a higher concentration of anthropogenic impact on agricultural land in Western Europe. This may be caused by the phosphorus fertilizers, which are historically of different origins in the western and eastern parts of the continent. While the Russian magmatic Kola

phosphate rock, the main source of P fertilizers in Eastern Europe, is practically free of Cd, that from Morocco, the main source of P fertilizer in Western Europe, contains Cd (Csillag et al., 2006).

3.4. Cobalt concentrations in topsoils of the European Union

Cobalt is an element which is essential to human health (e.g. it is part of vitamin B₁₂), but which in excess amounts can cause serious effects to lungs and heart (ATSDR, 2004a). It is worth noting that the transfer potential from soil to the edible parts of plants is rather low (Luo et al., 2010). The assessment of European soils revealed that excess cobalt is a real risk only in a few regions in Europe (Fig. S3B–D). While samples with concentration above the threshold value can be found in most European regions (Fig. S3A), these concentrations exceed the guideline

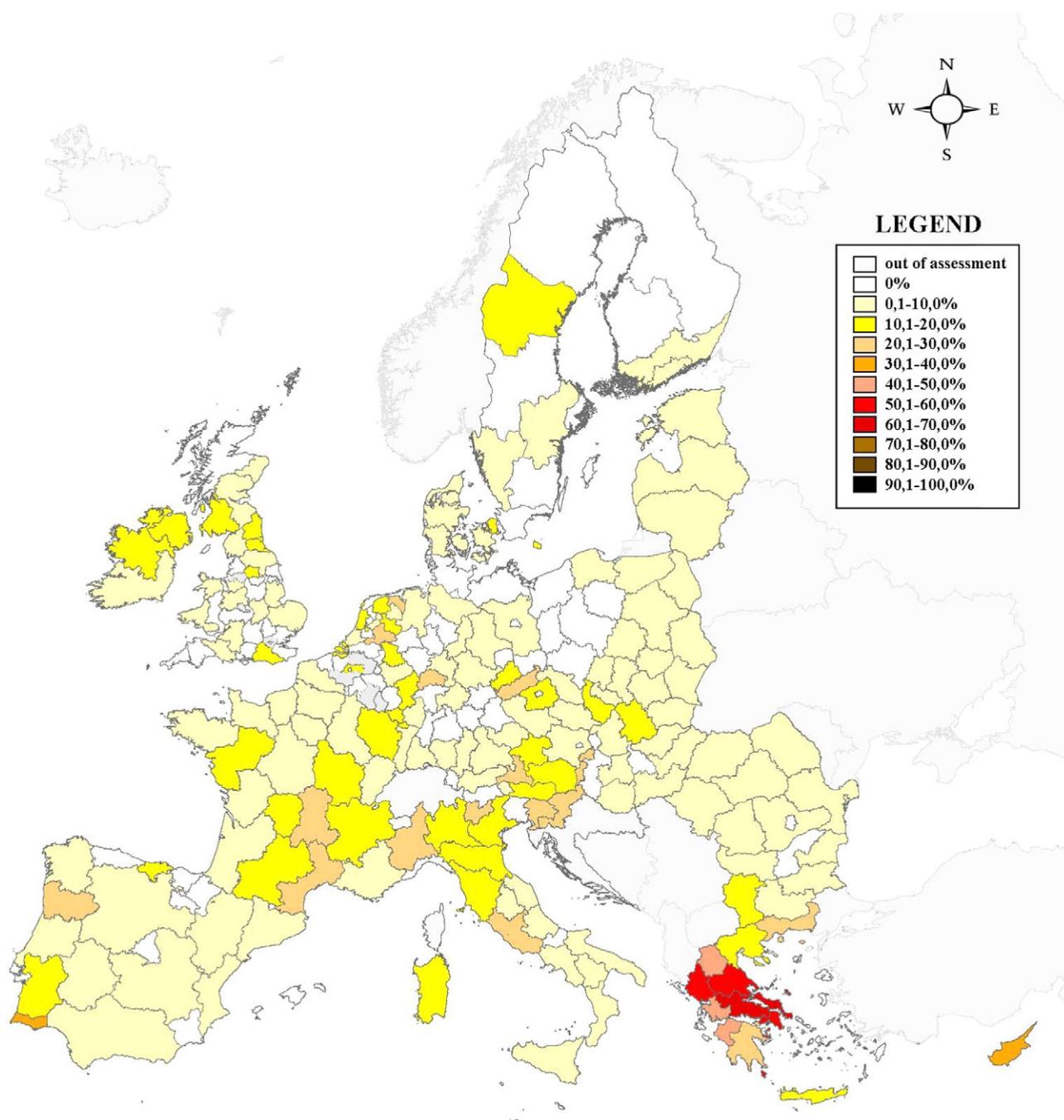


Fig. 4. Percentage of samples with concentrations above the lower guideline value in LUCAS samples from agricultural land.

values only in a few cases, both for agricultural and other land uses. One region in France and four smaller regions in Greece were found to have samples with cobalt concentrations above the guideline values for agricultural land (Fig. S3E). These samples represent a small percentage of the regions' total. However, while 4.5% of the samples from agricultural lands have cobalt concentrations above the investigation threshold, the guideline values are exceeded in 0.38% of the samples. This means that an estimated 668,000 ha of land are affected, which is five times the total agricultural land area of Luxembourg. Although cobalt is essential to life in a small amount and its deficiency has a negative effect on the human neurological system, exposure to higher doses of it can hamper lung functions. Thus a European policy should be adopted to monitor cobalt in soils.

3.5. Chromium concentrations in topsoils of the European Union

Chromium is quite abundant in most soils and 4.4% of all samples were above the threshold value. Although this figure is seemingly low, samples with concentrations above the threshold can be found in nearly half of the EU's NUTS2 regions (Fig. S4A). What is more noticeable, if we look at agricultural land is that 2.7% are above the threshold and 1.1% of the samples were above the guideline value (Fig. S4B, D). This figure shows that some 2 million ha agricultural land is at an ecological risk from high concentrations of chromium in soil. Our analysis shows that especially Piemonte, Lorraine-Alsace, Western-Macedonia and Central Greece are affected. As long term exposure to high Cr doses may cause adverse effects in the liver and the kidney and in situ remediation of

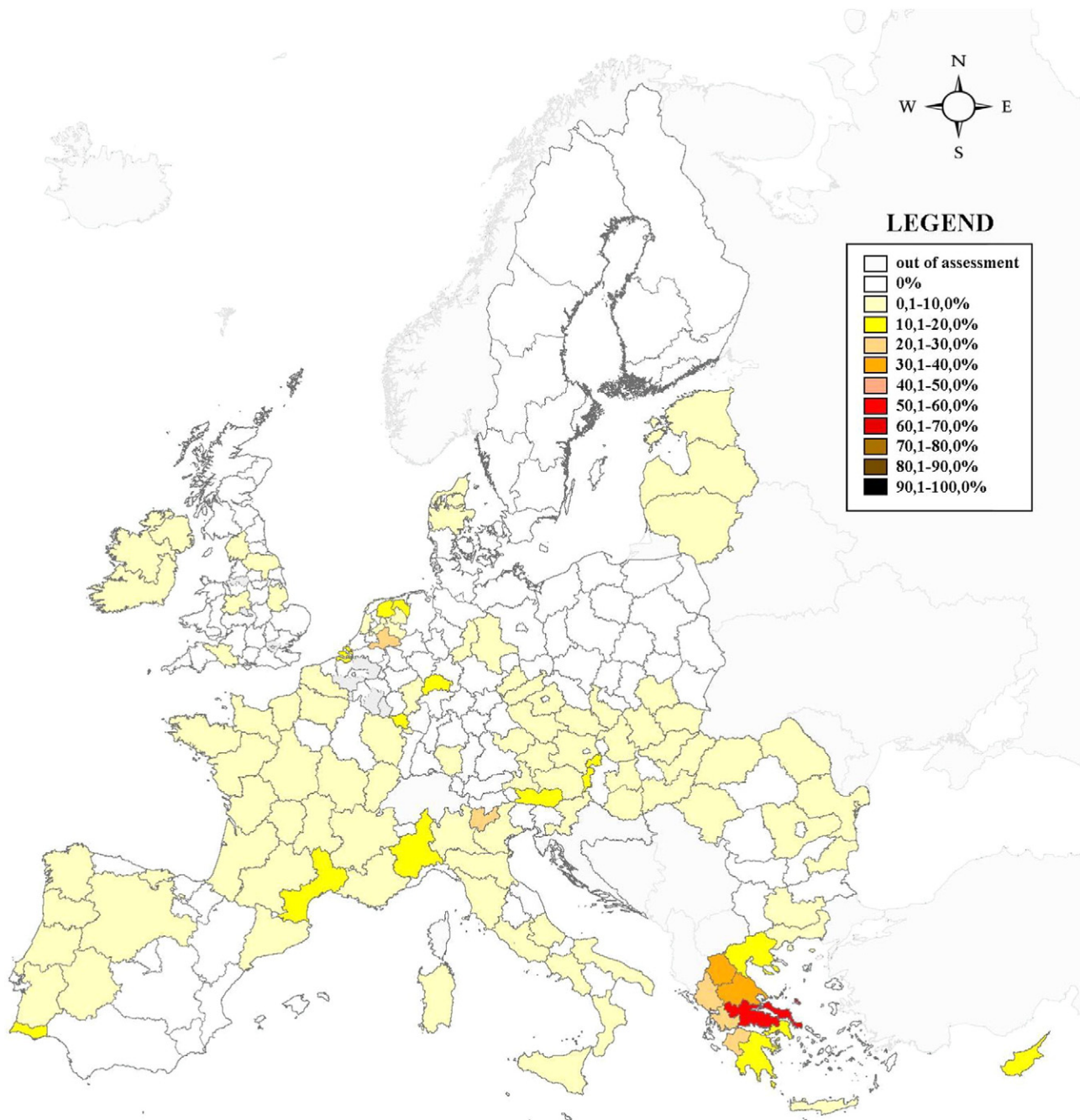


Fig. 5. Percentage of samples with concentrations above the higher guideline value in LUCAS samples from agricultural land.

soils affected by high concentration of chromium can be quite complicated (Palmer and Wittbrod, 1991; Pagilla and Canter, 1999), it would be especially important to take measures (e.g. controlling industrial sources) to prevent any further increase of Cr in the soil. Our study revealed the spread of chromium in agricultural land based on its elemental measures. However, it is worth noting that the highest risk arising from soil chromium is associated to its hexavalent form and trivalent Cr is relatively immobile in soil, thus present lower risk (McLean and Bledsoe, 1992).

3.6. Cu concentrations in topsoils of the European Union

Copper belongs to the substances which are essential for human health, e.g. by being part of enzymes involved in specific metabolic processes. However, it may be harmful in higher doses by causing

gastrointestinal distress, damage to liver, the immune system, neurological system and reproductive ability (ATSDR, 2004b).

Accumulation of copper in soils is mainly due to anthropogenic origin, such as mining or industrial activities, Agricultural use of products containing copper is also common, especially in pesticides applied in vineyards and orchards (Fishel, 2014). This might be a reason, why soil samples with high Cu concentrations can be found in the countries of the Mediterranean (Fig. S5A–F) where these land uses are common. Agricultural land is affected mostly in France, Italy, Portugal and Romania. Although the share of samples with Cu concentrations above the guideline value is rather low among all of the samples, its proportion exceeding 2% in some regions in France and Italy indicates a potential problem for food production. This is especially true, if we consider the higher proportion of samples with concentrations above the threshold value, which already indicates the presence of copper in a concentration

Table 2
Share of soil samples from the LUCAS survey with high concentrations of heavy metals.

Samples from agricultural land (n = 14,865)			
	Samples with concentrations above the threshold value	Samples with concentrations above the lower guideline value	Samples with concentrations above the higher guideline value
n	8632	928	380
%	58.07%	6.24%	2.56%
Samples from all land uses (n = 21,980)			
	Samples with concentrations above the threshold value	Samples with concentrations above the lower guideline value	Samples with concentrations above the higher guideline value
n	11,718	n.a.	523
%	53.31%	n.a.	2.38%

that requires precautionary thinking. Although most crops take up and accumulate Cu in small quantities only, continuous exposure to Cu in food may cause negative health effects in humans.

3.7. Hg concentrations in topsoils of the European Union

Mercury – either in inorganic forms or in its methyl compounds – may pose threat to kidney, liver the nervous-, and reproductive systems, as well as to the immune system. However, methylmercury species have higher bioavailability and toxicity. A recent study of the EFSA (2012b) suggests that inorganic mercury from the diet does not exceed the tolerable intake in Europe. Assessment of the LUCAS data confirms the very low risk posed by lead in agricultural soil in Europe. In fact, lead concentrations were found to exceed the threshold and the guideline values in a very small proportion (in 25 and 16 samples, respectively) of the samples from agricultural land. The affected samples originate from a few regions only. Historically, mining for gold and mercury leads to high Hg concentrations in mine areas. The latter may be the reason for the high Hg concentrations in some samples from Central Italy, North-West England and Eastern Slovakia (Fig. S6). Although these represent isolated cases only, the fact that some soil samples with Hg above the higher guideline values were still found on agricultural land of France, Germany, Italy and Spain calls for stricter control of Hg in all parts of the food chain, including soil. As mercury from human activities is the main source of Hg contamination (ATSDR, 1999; Steinnes, 1995) of soil today, contamination sources should also be under strict control.

3.8. Ni concentrations in topsoils of the European Union

Nickel in soil, like most other heavy metals may be of either natural or anthropogenic origin. While industrial activity may also be responsible for soil contamination in parts of Europe (Cempel and Nikel, 2006) according to the LUCAS survey there are also considerable differences between climatic regions regarding higher concentrations of Nickel in soil (Fig. S7). This fact suggests that soil Ni can be attributed to natural factors to a great extent. Our assessment suggests that the whole of the EU is affected by Ni contamination to some degree, but land under the influence of the last glaciation e.g. Germany, Poland and the Scandinavian countries are less prone to it. With the exception of the southern Apennine peninsula and most of Spain, samples with high concentrations of Ni can be found in considerable percentage of the samples from the Mediterranean region of Europe. The density of samples with Ni concentrations above the higher guideline value is the highest in Greece, where in some regions, more than half of the samples have Ni in higher concentrations (Fig. S7E, F). As excess amount of nickel may adversely affect the immune system as well as the reproductive system (ATSDR, 2005), our findings urges for more detailed analysis of soil nickel in the Mediterranean, even if the mobility and the potential

bioavailability of nickel is one of the lowest among heavy metals (Ma and Rao, 1997).

3.9. Pb concentrations in topsoils of the European Union

According to the data of the WHO (2015) the European population is the least prone to dietary lead intake. Exposure to lead from food materials in Europe is commonly much below the tolerable weekly intake, as the study of EFSA (2012c) reports. Exposure to lead occurs mainly through the food chain, although ingestion of soil and dust can also be an important contributor (EFSA, 2012c). Relatively low lead exposure can impair brain and nervous system – especially those of children – and elevated blood pressure, chronic kidney disease and probably cancer can be also caused by lead, even at relatively low blood lead levels (ATSDR, 2007a; IARC, 2006). Therefore a detailed assessment of the risk associated with lead in topsoil is required.

Based on our regional assessment, Central Italy, France Germany and the UK display the highest share of samples with relatively high concentrations of Pb in soils (Fig. S8A, B). Samples from the Baltic states, Finland and Hungary did not display detectable traces of lead contamination on agricultural land (Fig. S8B). The highest percentage of samples with Pb concentrations above the threshold value is found in Lazio province in Central Italy, probably due to the abundance of tertiary volcanic material in this region. However, none of these samples display a concentration above the guideline value for agricultural land. Such samples are very rare; only a few cases around Europe were found among the over twenty thousand samples, indicating that currently lead is not a problem for food safety. Nevertheless, the widespread occurrence of samples with concentrations above the threshold value, even if in small shares among the total number of samples, indicates the need for strict control of lead in agricultural land and – eventually – in the food chain.

3.10. Sb concentrations in topsoils of the European Union

Antimony may alter pulmonary function and may cause respiratory, neurological cardiovascular, gastrointestinal and hematological effects, as reported mostly based on data on exposure to airborne antimony dust (ATSDR, 1992). However, a study by Hammel et al. (2000) found correlation between the mobile fraction of antimony in the soils and antimony in leaves of spinach, proving that high concentration in soil can result accumulation of antimony in plants.

The LUCAS data shows that antimony is quite abundant in the topsoils in the European Union, with the highest density of samples with concentrations above the threshold value (both for agricultural areas and for all land use types) in Southern and Western Europe; Greece and Ireland, in particular (Figs. 1 and S9A, B). However, based on the application of the lower guideline value on the LUCAS soil samples, we can observe a much lower proportion of problematic samples and also with smaller areal coverage (Fig. S9C–F). In fact, while Greece and Ireland display antimony in most of their soil samples, the concentration of this element remains below the contamination threshold. Nevertheless, most of their areas require the assessment of their soil contamination and remediation need, just as most of Austria, Bulgaria, Catalonia, Northern Italy and Southern France. While precautionary measures seem to be necessary in these regions, and especially in Austria, France, Germany, Italy, Poland and Spain, where samples above the lower guideline values were found also on agricultural land, just a few samples were found in the whole EU with concentration above the higher guideline value. Although this result suggests that food of European origin is safe from Sb contamination, any further Sb load should be avoided, especially in some regions indicated above.

3.11. Zn concentrations in topsoils of the European Union

Zinc is an essential element for both plants and humans, but it is toxic in excess amounts (Swartjes, 2011). Therefore it is crucial to

control its right amount in agricultural soils. It may have direct toxic effects and, among others, may cause gastrointestinal and immunologic problems. In the same time, high amounts of Zinc probably also inhibit copper absorption, thus resulting copper deficiency symptoms. Zinc is propagated throughout the food chain by bioaccumulation resulting higher concentrations in meat products than in vegetables and fruits (ATSDR 2007b). While zinc deficiency can be attributed to soil factors like high pH, high Ca and CaCO₃ concentrations (Alloway, 2008), its excess in soil might be either of geological or anthropogenic origin. The LUCAS data shows that excess zinc appears in agricultural land in more than 20% of the NUTS regions in Europe by showing concentrations above the threshold concentration (Fig. S10A, B). However, the percentage of these samples in the total is less than half of 1% (0.004500/0). The number of samples in which the zinc concentration exceeds the higher guideline value is just above a dozen out of the more than twenty thousand in total. We can state that zinc pollution exists only in isolated cases in the agriculture of the EU. Consequently, this metal presently does not hold any risk to food safety on a continent al scale. However, it is worth noting that different zinc species are absorbed at different rates, which may result different risk of toxicity depending on the local conditions.

4. Conclusions

Data from the LUCAS Topsoil Survey shows that the immense majority of European agricultural land can be considered adequately safe for food production. However, based on the continent-wide survey the high share of samples with HM concentrations above the assessment threshold indicate large areas where precautionary measures are needed. At the same time, an estimated 6.24% of the agricultural land needs local assessment and eventual remedial action, based on the guideline concentrations applied in our study. Although at first sight this percentage of land with soils of relatively high concentrations of heavy metals does not seem to be alarmingly high, the real areal coverage of this low percentage share can total 137,000 km² of agricultural land in the European Union. The survey-based evidence on the areal extent of this problem should lead to relevant policy action on the remediation and management of soil resources across Europe. Preventive measures applied at the critical lands to exclude harmful substances from the food chain can include the control of bioavailability of the elements (e.g. by liming or applying other methods for demobilizing heavy metals), but also production of materials other than foodstuff on affected land, and eventually also remediation actions.

The results of the assessment based on the soil samples from the LUCAS survey also highlight the need for spatially intensified and thematically broadened monitoring of soil resources in the European Union. We also suggest to establish harmonized screening values for soil contamination in the EU, to gain better understanding of the magnitude and details of the problem associated with heavy metals in soil. Such, harmonized system could provide multiple benefits for planning the sustainable utilization of land resources in the European Union.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <http://dx.doi.org/10.1016/j.envint.2015.12.017>.

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