Science of the Total Environment 565 (2016) 1054–1062

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Maps of heavy metals in the soils of the European Union and proposed priority areas for detailed assessment

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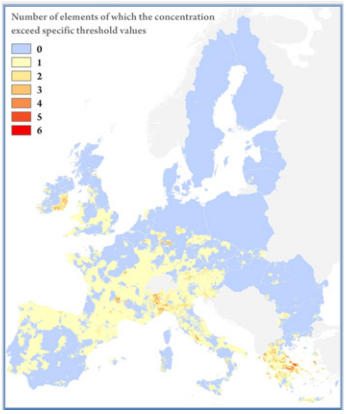
HIGHLIGHTS

- Detailed maps of heavy metals in the topsoil of the European Union are presented.
- As, Cd, Cr, Cu, Hg, Pb, Zn, Sb, Co and Ni concentrations in European topsoil is mapped.
- Priority areas where potential threat from heavy metals are high are delineated.
- 1.2 M km^2 or 28.3% of the total surface area of the EU is proposed for further assessment.
- Historical and recent industrial and mining areas show elevated As, Cd, Pb and Hg levels.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 25 March 2016
Received in revised form 12 May 2016
Accepted 17 May 2016
Available online 31 May 2016

Editor: F.M. Tack

Keywords:
Soil contamination
Soil threats
As, Cd, Cr, Cu, Hg, Pb, Zn, Sb, Co, Ni

ABSTRACT

Soil contamination is one of the greatest concerns among the threats to soil resources in Europe and globally. Despite of its importance there was only very course scale (1/5000 km^2) data available on soil heavy metal concentrations prior to the LUCAS topsoil survey, which had a sampling density of 200 km^2. Based on the results of the LUCAS sampling and auxiliary information detailed and up-to-date maps of heavy metals (As, Cd, Cr, Cu, Hg, Pb, Zn, Sb, Co and Ni) in the topsoil of the European Union were produced. Using the maps of heavy metal concentration in topsoil we made a spatial prediction of areas where local assessment is suggested to monitor and eventually control the potential threat from heavy metals. Most of the examined elements remain under the corresponding threshold values in the majority of the land of the EU. However, one or more of the elements exceed the applied threshold concentration on 1.2 M km^2, which is 28.3% of the total surface area of the EU. While natural backgrounds might be the reason for high concentrations on large proportion of the affected soils, historical and recent industrial and mining areas show elevated concentrations (predominantly of As, Cd, Pb and Hg) too, indicating the magnitude of anthropogenic effect on soil quality in Europe.

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http://dx.doi.org/10.1016/j.scitotenv.2016.05.115

1. Introduction

Soil contamination is one of the greatest concerns among the threats to soil resources in Europe and globally (CEC, 2006; Kong, 2014). Heavy metal, together with mineral oils, is the most frequent contaminant in European soil. Recognizing the importance of this problem and consequent need to halt further contamination and start cleaning the soil of the European Union (EU), the 7th Environment Action Programme (OJEU, 2013) of the EU sets the aim to ensure that by 2020 "soil is adequately protected and the remediation of contaminated sites is well underway". A recent study by van Liedekerke et al. (2014) estimates the number of potentially contaminated sites in Europe to sum up to 2.5 Million, illustrating the extent of this challenge. While the new inventory of polluting sites by van Liedekerke et al. (2014) reflects on the magnitude of the problem with regards to risk from point sources, to date no detailed information was available on the size of the area affected by soil contamination in Europe. The FOREGS data produced by the EuroGeoSurvey (Salminen et al., 2005) and the derived continuous map sheets (Lado et al., 2008) have been the most comprehensive sources of information in this respect so far. However, the low sampling density (1 site/5000 km^2) of the FOREGS study (Demetriades et al., 2010) limits the possibilities of interpretation of the results to continental scale overview only.

The LUCAS Topsoil Survey of the European Union (Tóth et al., 2013) opened new possibilities to acquire detailed information on the soil cover in Europe, including heavy metal content. With its 1 site/200 km^2 sampling density it allows modeling and monitoring of soil resources on a finer scale than any previous attempts. This detail of sampling is adequate to create continuous maps for reliable spatial representation at 1 km resolution (Hengl, 2006) of heavy metals in topsoil of Europe. Comparing this detail in information with the sampling density of the geochemical and mineralogical survey of the United States Geological Survey, which was 1 site/1600 km^2 and resulted maps of heavy metals in the US (Smith et al., 2014), the current data can be regarded as a major step in the appraisal of heavy metal in European topsoil. Further to providing information for regional comparison, the LUCAS topsoil data offers the first appraisal for countries where national inventories did not exist before, or supplement the information in countries where monitoring sites are in low density (Jones et al., 2005).

Using this new survey data and applying fully validated comprehensive geostatistical method our aim was to produce the most detailed and up-to-date coverage of heavy metal in the topsoil of the European Union. Based on the maps of heavy metal concentration in topsoil we made a spatial prediction of areas where local assessment is suggested to monitor and eventually control the potential threat from heavy metals.

While heavy metal concentrations are predicted for topsoil of the whole territory of the European Union, differentiation between natural backgrounds and anthropogenic pollution was not the aim of the current research.

2. Materials and methods

2.1. Databases used

2.1.1. The LUCAS topsoil database

Over 23,000 topsoil samples (upper 20 cm) were collected from land of the European Union (EU) Member States (EU-28 except Croatia) with the aim to produce the first coherent baseline topsoil database for continental scale monitoring (Tóth et al., 2013). Sampling was performed in two campaigns, in 2009 and in 2012. The soil sampling was undertaken within the frame of the Land Use/Land Cover Area Frame Survey (LUCAS), an EU wide project to monitor changes in the management and character of the land surface (Eurostat, 2015). The survey, which represents the first effort to build a consistent spatial database of soil properties for environmental assessments is applied standard sampling and analytical procedures.

A stratified sampling scheme was applied and samples were taken from all land cover classes with unique georeferenced locations (Tóth et al., 2013). Basic soil properties, such as particle size distribution, pH, organic carbon, carbonates, NPK, cation exchange capacity (CEC) and multispectral signatures were determined from each sample, which were also analysed for heavy metal content. Analysis of soil parameters followed standard procedures. Tóth et al. (2013) provided detailed description on the methodology and data of the LUCAS Topsoil Survey.

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, Hg, Ni, Pb, Sb and Zn were carried out. Elements were analysed 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 analysed using the prEN16174 (ECS, 2010) procedure. The unit of measurement was mg/kg for As, Cd, Cr, Cu, Hg, Pb, Zn, Sb. Co and Ni, with detection limits 2.84, 0.07, 0.32, 0.26, 0.00005, 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.

2.1.2. Auxiliary data

The applied environmental co-variables consisted of topography, geology, vegetation, meteorology, as well as anthropogenic factors.

2.1.2.1. Topography. Topography was taken into consideration by the EU-DEM dataset, which is one of the most detailed, freely available Digital Elevation Models. The EU-DEM is a 3D raster dataset with elevations captured at 1 arc second postings or about every 30 m. From EU-DEM we got the following 18 derivatives by SAGA GIS (Bock et al., 2007): (1) Aspect, (2) Channel Network Base Level, (3) Closed Depressions, (4) Convergence Index, (5) Cross Sectional Curvature, (6) Longitudinal Curvature, (7) Diurnal Anisotropic Heating, (8) Elevation, (9) LS Factor, (10) Mass Balance Index, (11) Multiresolution Index of Ridge Top Flatness (MRRTF), (12) Multiresolution Index of Valley Bottom Flatness (MRVBF), (13) Slope, (14) Topographic Wetness Index, (15) SAGA Wetness Index, (16) Total Direct Solar Insolation, (17) Valley Depth, and (18) Vertical Distance to Channel Network.

2.1.2.2. Geology. On a spatial basis, the lithogenic source (geological minerals in the soil parent material, on which the soil has developed) is the dominant factor determining the total concentration of heavy metals in world soils (Alloway, 2013). Geology data originated from the 'Digital data of the 1:5 Million International Geological Map of Europe and Adjacent Areas' (IGME 5000, Asch, 2005). The raw map with its 100 different rock types would be too detailed for the mapping, therefore the types were merged by Bakacsi (2016) into 16 categories according to their chemical properties: (1) igneous acidic, (2) igneous intermediate, (3) igneous basic, (4) igneous ultrabasic, (5) sedimentary calcareous, (6) sedimentary siliclastics, (7) sedimentary undifferentiated, (8) metamorphic acidic, (9) metamorphic intermediate, (10) metamorphic basic, (11) metamorphic ultrabasic, (12) metamorphic calcareous, (13) metamorphic siliclastics, (14) metamorphic undifferentiated, (15) volcano-sediments intermediate, and (16) volcano-sediments undifferentiated.

2.1.2.3. Vegetation. State of vegetation can especially be related to the heavy metal concentration in soils. Like all living organisms, plants are often sensitive both to the deficiency and to the excess availability of some heavy metal ions as essential micronutrient, while the same at higher concentrations and even more ions such as Cd, Hg, and As are strongly poisonous to the metabolic activities (Nagajyoti et al., 2010). Agricultural areas are particularly affected by heavy metal contamination due to the direct applications of organic or inorganic fertilisers, and agrichemicals (Alloway, 2013). Vegetation was taken into consideration by remotely sensed images, since MODIS enhanced vegetation index (EVI) effectively characterizes bio-physical/ biochemical states and processes from vegetated surfaces. Principal Components (2005–2006), furthermore mean value of the monthly MODIS EVI time series data (2001 − 2012) were used in the mapping process (MODIS, 2014).

2.1.2.4. Climate. Natural and man-made processes have been shown to result in metal containing airborne particulates. Depending on prevailing climatic conditions, these particulates may become wind-blown over great distances; nonetheless, they are subjected to the fate that they are ultimately returned to the lithosphere as precipitations by rain or snowfall (Nagajyoti et al., 2010). Climatic parameters can be described by various different spatial datasets. In the mapping process we used the following 10 variables from two different databases: (1) Annual mean temperature, (2) Mean diurnal range, (3) Isothermality, (4) Temperature seasonality, (5) Maximum temperature of the warmest month, (6) Minimum temperature of the coldest month, (7) Temperature annual range, (8) Mean temperature of the warmest quarter, (9) Mean temperature of the coldest quarter (EuroLST BIOCLIM: Metz et al., 2014), and (10) Sum of average monthly precipitation (WorldClim – Global Climate Data).

2.1.2.5. Human impact. Anthropogenic factors cannot be omitted from mapping heavy metal distribution in soil. Human impact – aside from agriculture, which was mentioned previously – is concentrated near cities, roads and industrial areas. Spatial distribution of the persistent lights at night trace well out these techno-areas. The layer is obtained from the Defense Meteorological Satellite Program.

2.2. Methods applied

2.2.1. Data preparation

Exploratory data analysis was performed on the LUCAS dataset to identify the outliers and examine the marginal distribution of the heavy metals, respectively. All of the marginal distributions have shown a positively skewed distribution, which is common in the case of earth and environmental science data (Manchuk et al., 2009). In fact, most of the geostatistical techniques rely on the MultiGaussian (or multivariate normal) approach. Hence, all of the observations on heavy metals have been transformed to their natural logarithm. The transformed values show a symmetric marginal distribution close to normal. The outliers were identified and then removed from the dataset.

A control dataset was created, which was independent from the mapping procedure. The control dataset incorporated 4396 randomly selected observation points, which is approximately 20% of the LUCAS dataset. The size of the control dataset is adequate to make statistical inferences for the accuracy of the compiled heavy metals maps.

Covariate data layers were resampled in SAGA GIS into a common grid system with 1 km resolution and all further processing was performed on this dataset.

2.2.2. Geostatistical prediction method

Regression kriging (RK) spatial prediction method was used to map the heavy metals spatial distributions. The RK technique combines the regression of the dependent variable on environmental covariates with kriging of the regression residuals. The estimation for z variable at an unvisited location $\mathbf{s}_0$ is:

$$z(\mathbf{s}_0) = \mathbf{q}_0^{\mathbf{T}} \cdot \beta + \boldsymbol{\lambda}_0^{\mathbf{T}} \cdot (\mathbf{z} - \mathbf{q} \cdot \beta)$$

where β is the vector of the regression coefficients, $\mathbf{q}_0$ is the vector of the covariates at the unvisited location $\mathbf{s}_0$, $\boldsymbol{\lambda}_0$ is the vector of the kriging weights, $\mathbf{z}$ is the vector of the observations and $\mathbf{q}$ is the matrix of the covariates at the sampling locations. The prediction error variance of RK at $\mathbf{s}_0$ is given by:

$$\sigma^2(\mathbf{s}_0) = c(0) - \mathbf{c}_0^{\mathbf{T}} \cdot \mathbf{C}^{-1} \cdot \mathbf{c}_0 + \\ + \left(\mathbf{q}_0 - \mathbf{q}^{\mathbf{T}} \cdot \mathbf{C}^{-1} \cdot \mathbf{c}_0\right)^{\mathbf{T}} \cdot \left(\mathbf{q}^{\mathbf{T}} \cdot \mathbf{C}^{-1} \cdot \mathbf{q}\right)^{-1} \cdot \left(\mathbf{q}_0 - \mathbf{q}^{\mathbf{T}} \cdot \mathbf{C}^{-1} \cdot \mathbf{c}_0\right),$$

where $c(0)$ is the variance of the residuals, $\mathbf{c}_0$ is the vector of covariances between the residuals at the observed and unvisited locations and $\mathbf{C}$ is the covariance matrix of the residuals.

2.2.3. Validation method

Locally the accuracy of the prediction was estimated by the prediction error variance. Overall accuracy of the created heavy metals maps was evaluated using the control dataset. The following indicators were derived using the predicted $z(\mathbf{s}_i)$ and observed $z(\mathbf{s}_i)$ values of the heavy metals at the control points $\mathbf{s}_i$:

$$\mathrm{ME} = \frac{1}{n} \cdot \sum_{i=1}^{n} [z(\mathbf{s}_i) - z(\mathbf{s}_i)]$$

$$\mathrm{MAE} = \frac{1}{n} \cdot \sum_{i=1}^{n} |z(\mathbf{s}_i) - z(\mathbf{s}_i)|$$

$$\mathrm{RMSE} = \sqrt{\frac{1}{n} \cdot \sum_{i=1}^{n} [z(\mathbf{s}_i) - z(\mathbf{s}_i)]^2}$$

where n is the number of the control points (in this case 4396). ME (mean error) is often referred to as the bias. RMSE (root mean square error) quantifies the spread of the errors distribution, where the error is defined as the difference between the predicted and observed values (Isaaks and Srivastava, 1989). A reasonable goal for any mapping procedure is to produce unbiased predictions with small spread. MAE (mean absolute error) provides information about the average absolute error, which is more rigorous than ME, according to its definition.

2.2.4. Principal component analysis of heavy metal maps

In order to analyze complex linkages between different concentrations of heavy metals in their geographical context, principal component analysis (PCA) was performed. Because the concentration of some elements have gradual changes while that of others have sharper changes throughout the landscape of Europe the results of the PCA represent the sum of conditions, expressed by the main factors in the system. Results of the PCA analyzes are also mapped to indicate areas with these established interactions.

2.2.5. Application of threshold values to identify risk areas

Results of the mapping exercise performed in our study enables the identification of areas where heavy metal accumulation presents a hazard to health or the environment. To delineate those areas which are threatened by high heavy metal concentrations, we applied assessment thresholds on each map.

Threshold values proposed by the Finnish Ministry of Environment (MEF, 2007) were used, which indicate concentrations where soil contamination and remediation needs must be further assessed. The threshold values of the FEM (2007) represent a good approximation of the mean values of different national systems in Europe (Carlon et al., 2007), have been used in an international context for agricultural soils (UNEP, 2013) and were also successfully applied in regional assessment in Europe (Tóth et al., 2016). Soil covers with element concentration above the thresholds are displayed regardless of the origin of the given substance. Naturally occurring normal concentrations (background concentrations) or raised concentrations of hazardous substances that are found in topsoil which are suspected to be contaminated are treated equally in our delineation.

3. Results and discussion

3.1. Results of the geostatistical predictions

Nested models were used throughout the variogram modeling, which combine two or more basic variograms to model the spatial variability (Goovaerts, 1997). In this study three basic structures were used, i.e. a nugget effect and two exponential models. The first one arose from the measurement errors and/or small-scale heterogeneity, while later ones were applied to model the short range (10–100 km) and long range (300–1000 km) spatial variability. The short range variability can be considered as local and regional scale heavy metals distributions (e.g. industrial or mining activities, geological formations), while the long range spatial variability can represent continental scale tendencies.

Topsoil heavy metal concentrations show skewed distribution for all of the elements examined, with outliers of high concentrations. The number of outliers, which probably indicate polluted sites with point source pollution was the highest in the cases of Cd, Cu, Cr, Pb, Hg and Sb. An overview of the main statistics on heavy metal content predicted by the maps produced is given in Table 1.

The maps of Fig. 1 (a–i) show the concentrations of various heavy metals in the topsoil of the European Union. Heavy metal in topsoil is either of natural origin or from anthropogenic sources. Human activities can result pollution of limited areas with well-defined boundaries from point sources or diffuse contamination of larger land surfaces. The recognition of the origin of heavy metal is often not very easy, if the point source cannot be identified at the location where high concentration of element is found. The maps of heavy metal content of topsoil with European coverage help to assess both the spatial tendencies and hotspots in the continent.

3.2. Geographical tendencies of heavy metals in topsoil of the EU

Results of the PCA analysis (Table 2.) indicate close correlation of Co, Cr, Cu, Mn, Ni and Sb, which are included to the first PC describing nearly half of the total variance and among which both siderophiles and chalcophiles elements can be found. PC-2, adds another 16% to the explanation of the variance in the system, by its main constituents, As, Cd, Hg and Pb, which are chalcophile elements. With PC-3, including Cd and Pb, two elements which likely to indicate anthropogenic pollution, the total explained variance reaches 75%.

Table 1
Summary statistics of heavy metal concentrations.

Element	Concentration (mg/kg)			
Name	Min	Max	Mean	Std. deviation
Cd	0.02	3.17	0.09	0.11
As	0.46	252.53	3.72	2.92
Co	0.32	91.89	6.35	4.3
Cr	1.57	273.94	21.72	15.7
Cu	0.91	159.07	13.01	9.4
Hg	0	1.59	0.04	0.04
Mn	9.62	2285.23	373.05	237.68
Ni	0.36	466.48	18.36	18.15
Pb	1.63	151.12	15.3	8.33
Sb	0.01	10.91	0.25	0.37

When displaying the PCs on maps (Fig. 2a–c) we can see that the majority of interactions in PC-1 occur in the Central and Eastern Mediterranean region. Further contacts integrated to PC-1 are abundant also in other parts of the continents where glaciation did not interrupt soil genesis, except for the Carpathian basin and the Iberian peninsula, which regions have low concentration of these elements in their topsoils. PC-2, including four chalcophile elements is characteristic for the north-western part of the continent and PC-3, with Cd and Pb appears nearly all parts, although on limited areas, except for those abundant appearances in Ireland and Greece.

The examinations by elements show that <20 mg/kg As was detected in >95% of LUCAS topsoil samples and high portion of the samples contain Arsenic below the limit of detection (LOD). As is most abundant in topsoil of mountainous areas, such as the Alps, the Carpathians, the Massif Central or the Pyrenees (Fig. 1a). Alluvial As deposition can be observed in higher concentration in Holocene deposits at the foothills of younger mountains, where fluvial redistribution may also occur. The Po plain is a prominent example for such a process (Bianchini et al., 2014). On the other hand, post-glacial earth material of quaternary origin is practically free of As, as can be observed at the Northern European regions.

Cd levels in European soils can be characterized by irregular distribution throughout the continent (Fig. 1b.). The largest region with high concentration of Cd can be found in the limestone shale belt of Ireland, which finding is in accordance with the results of local survey (McGrath and Fleming, 2006). Bavarian Alps and the South Eastern Alps also show geographical tendencies of elevated Cd in topsoil. Based on the LUCAS topsoil data low Cd concentrations are predicted for Bulgaria, Portugal and the Scandinavian country. The Carpathian basin is practically free of Cd.

The major geological divide between topsoils of different Co concentration is the limit of the last glaciation (Fig. 1c.). The Co concentration in the postglacial surfaces of Northern Europe is rather low. Older surfaces south of the 55° latitude, where longer periods of soil formation allowed higher rates of mineral transformations exhibit higher Co concentrations. Higher Co concentrations can be observed also in areas associated with mountainous terrain, the Apennines and the Carpathians in particular, but Co concentrations in these areas are still not significantly different from other parts of the continent. High concentrations in the Massif Central in France, the Pindus Mountains in Greece and the Tatras in Slovakia are isolated cases where high concentrations can be observed on a regional scale, due to geochemical reasons.

The boundary of recent postglacial area north of the 50° latitude is the major divide also for Cr content in topsoil in different regions on a continental scale (Fig. 1d). There are six regions with predominantly geology-derived Cr accumulation, of which two has relatively large areas involving agricultural land uses. One in Central to North-Western Greece and the other in Piemonte and the Po plain in Northern Italy.

Cu, like many other elements can be found only in small, but still detectable concentrations in the postglacial plains and somewhat higher, but still low concentrations in the mountainous land of Northern Europe (Fig. 1e.). On the contrary, the Mediterranean hosts high concentrations of Cu in topsoil, the Apennine peninsula, in particular. Croplands contain Cu in significantly higher concentrations than woodland or in this region. When comparing our results with the map of Lado et al. (2008) we can see both similarities and differences. For instance Northern Italy and Andalusia are found to be affected by higher concentration in both maps, but the assessment based on the LUCAS shows generally higher Cu levels in these regions than what was previously predicted based on the FOREGS data.

Hg of higher concentrations in European topsoil can mainly be found in the Eastern ranges of the Alps and the Northern Carpathians and in Lazio province around Rome in Italy (Fig. 1f). The LUCAS based maps partly confirm the finding of the FOREGS analysis (Ottesen et al., 2013)

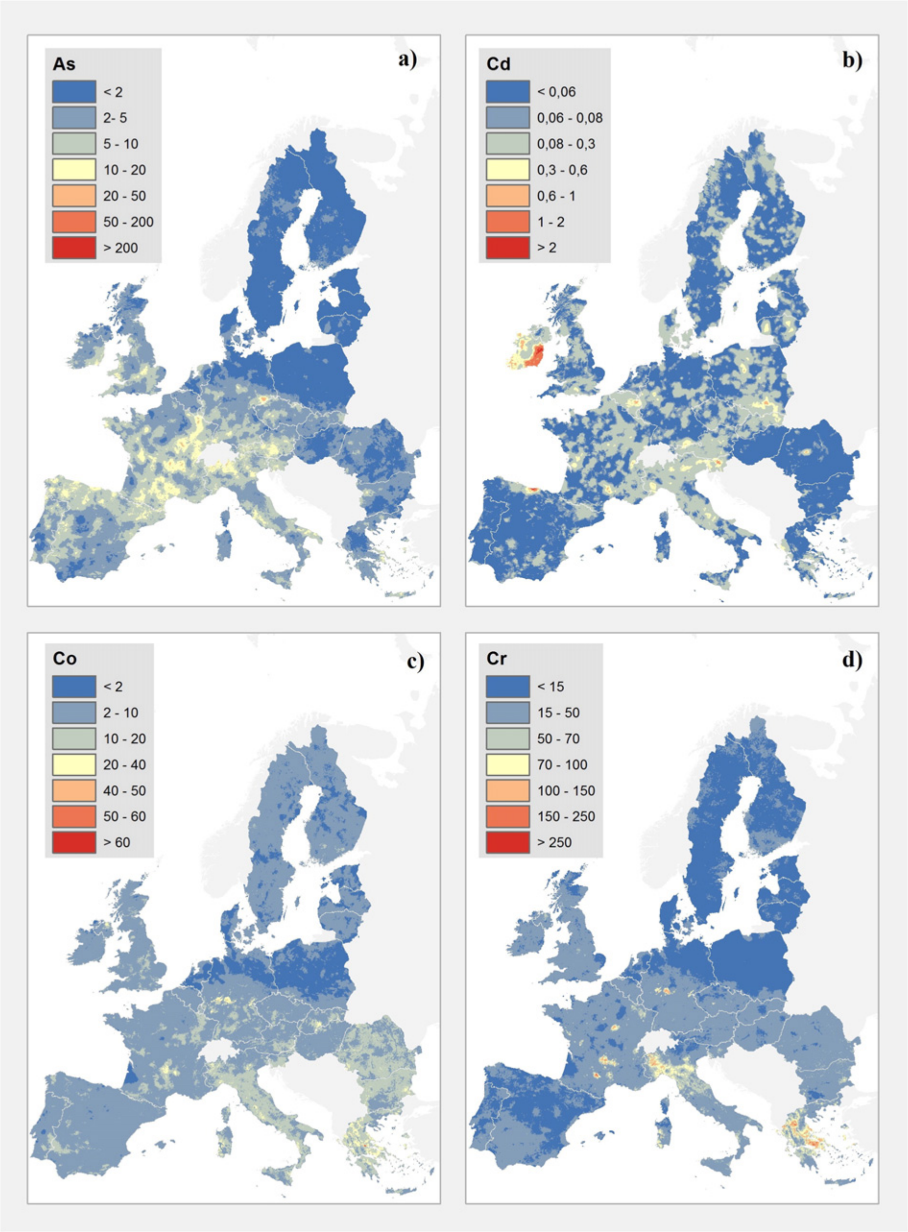

Fig. 1. Concentration of heavy metals in topsoil of the European Union.

in that the continental-scale distribution of the element is dominated by geology. On the other hand, some hotspots with high Hg concentrations of human release are clearly visible on the LUCAS based continental maps (for details see Section 3.3). The climate-based comparison did not confirm significant difference between main climate zones of Europe.

Mn concentration in European topsoil shows irregular geographic distribution (Fig. 1g). Only the postglacial northern areas contain low concentration while on other parts of the continent Mn is generally more abundant in topsoil. The Southern Sub-Continental areas as well as Central- and Eastern Mediterranean have most diverse Mn contents, including regions with mean Mn concentrations above 1000 mg/kg soil.

Ni is among the elements of which the concentration in topsoil is geographically divided by the last glaciation (Fig. 1h). North of the 55° latitude its concentration is generally low, although in the Baltic States some 5% of the samples were found to have Ni concentration above 100 g/kg. This share is comparable with that of whole EU where over 95% of areas in the EU contain <100 mg/kg Ni. It is worth mentioning that, according to the MEF (2007) this is the guideline value for ecological risk assessment. The highest concentration areas for Ni are in the Piemonte region in North-Western Italy and to Northern Greece, including the northern part of the Peloponnesus.

The major geological divide of the last glaciation has a smaller influence on topsoil lead concentration than in the cases of other heavy metals (Fig. 1i), probably because this element is mostly of anthropogenic origin in topsoil. High levels of Pb concentrations can be found in many regions of Europe, but those have the highest where historical industrial activities were combined with mining. Such cases are evident in southern Saxony, Central Saxony-Anhalt and North Rhine-Westphalia (Bergisches Land) in Germany, around Bristol and Manchester in England and Rome, Italy.

Sb is abundant in most European topsoil (Fig. 1j), especially in the southern and western part of the continent. Ireland and Greece have particular high concentrations of Sb in the upper layer of their soil cover but Sb accumulation ca also be seen in the North-Eastern Alps,

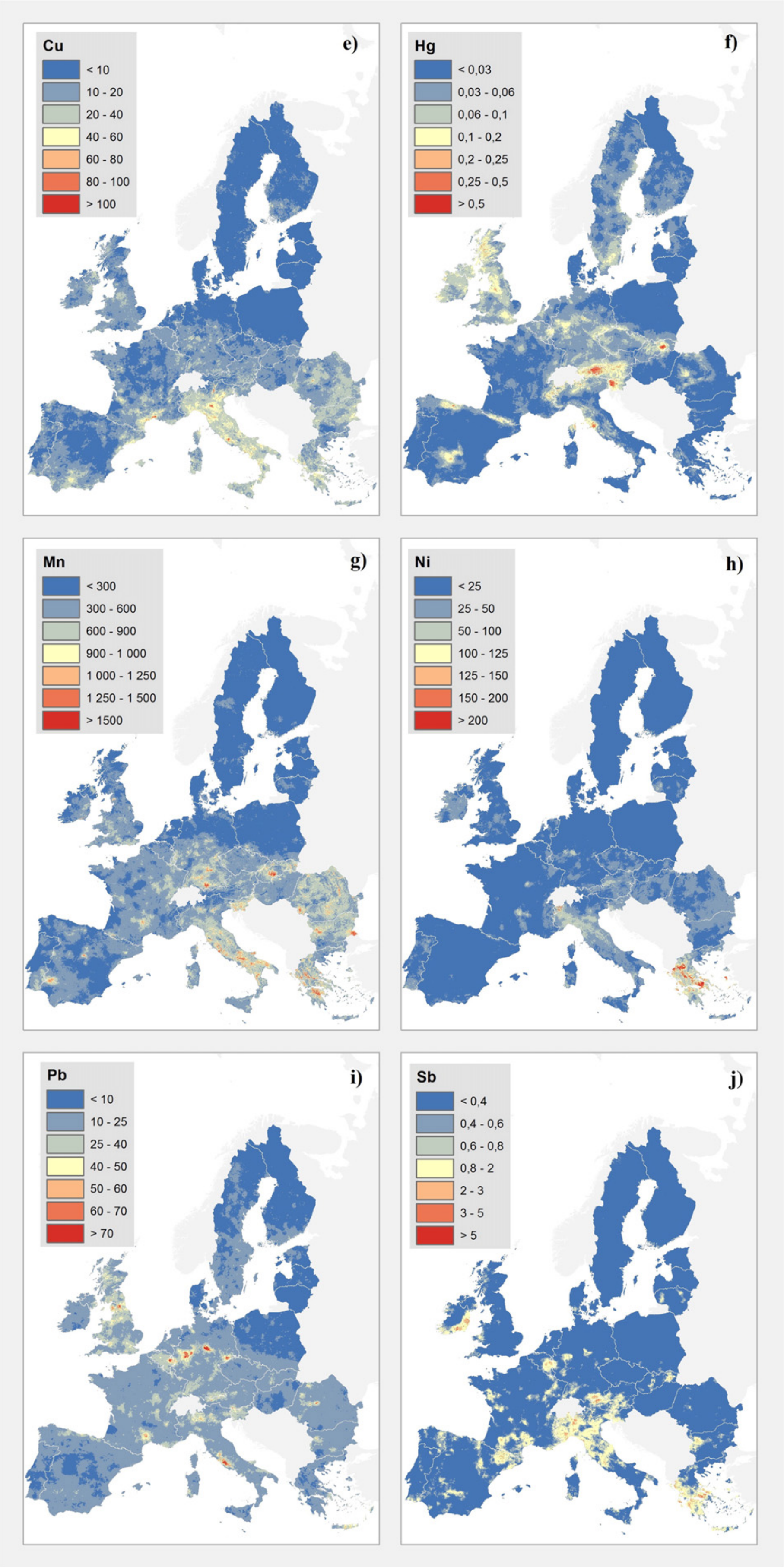

Fig. 1 (*continued*).

Table 2
Results of the principal component analysis on 10 heavy metals (gray shadings indicate main constituents of the PCs).

	PC–1	PC–2	PC–3	PC–4	PC–5	PC–6
As	−0.255	−0.335	0.183	−0.662	0.215	0.464
Cd	−0.110	−0.446	−0.682	0.020	−0.530	0.108
Co	−0.398	0.245	0.097	0.055	−0.157	0.173
Cr	−0.394	0.252	−0.067	0.111	0.104	0.196
Cu	−0.375	0.113	0.021	−0.088	−0.047	−0.646
Hg	−0.131	−0.531	0.253	0.657	0.206	0.119
Mn	−0.376	0.153	0.159	−0.015	−0.461	0.075
Ni	−0.358	0.235	−0.250	0.291	0.252	0.289
Pb	−0.282	−0.377	0.442	−0.016	−0.232	−0.236
Sb	−0.320	−0.222	−0.373	−0.144	0.513	−0.358
Explained variance (%)	48.72	16.19	9.79	7.55	5.32	4.19
Explained cumulative variance (%)	48.72	64.91	74.71	82.26	87.58	91.77

in the region from Wallonia to the Ruhr land, in North-East Italy, Slovakia and the South Pyrenees.

3.3. Hotspots and delineation of priority areas for monitoring

After centuries of anthropogenic emission of heavy metals to the environment it is quite difficult to differentiate natural background contents of heavy metal from diffuse pollution in many cases. Background concentrations can be assessed against polluted sites where the source can be identified (FDEP, 2012), but the methodology to determine natural background concentrations of polluting substances are not straightforward and results might be biased especially in urban or industrial districts (Ander et al., 2013). The task to define areas where high concentrations of heavy metal in topsoil should be regarded as pollution was beyond the scope of the current study. However, those hotspots where the threat of high level of heavy metal is present can be identified based on the maps presented in this paper. Either. While the differentiation between natural and anthropogenic heavy metal and the detection of the magnitude of anthropogenic effect was out of the scope of our current study, some observations on areas with high concentration can be made based on the maps produced.

As seems to be one of the major threats to the topsoil in Southern Saxony which is one of the main historical mining areas in Germany (Fig. 1a). Cd is present in distinct areas in the Cantabria region in Northern Spain, a traditional Zn mining area where sandy conglomerates are combined with dolomite (ITGE, 1994) and also in the Ruhr region of Germany, where high concentration of Co and other heavy metals are also found, around Lyon and Nimes in France and southern Poland, which areas are all home of heavy industry and mining (Fig. 1b). The Mures county in Central Rumania was also an important mining district for centuries. Historically, mining for gold and mercury leads to high Hg concentrations in those mine areas. This may be the reason for the high Hg concentrations in some samples from Central Italy, North-West England and Eastern Slovakia (Fig. 1f).

All the above areas are included to the list of priority regions where more detailed assessment is proposed (Fig. 3). Based on the appraisal of risk areas under threat of soil contamination in Europe, most of the Western-Central European region, Greece, Central Italy and South East Ireland should be examined more in detail. Monitoring of the evolution of heavy metal contents in soil and related ecological systems, including crops is proposed on these lands, which cover more than a quarter of the land surface of the European Union (Table 3.). In order to identify anthropogenic pollution and natural background concentrations, subsoil and topsoil heavy metal concentrations should be compared. As currently such data is not available for reliable assumptions in the required density for Europe, we propose to extend the future LUCAS surveys towards the assessment of subsoil quality as well.

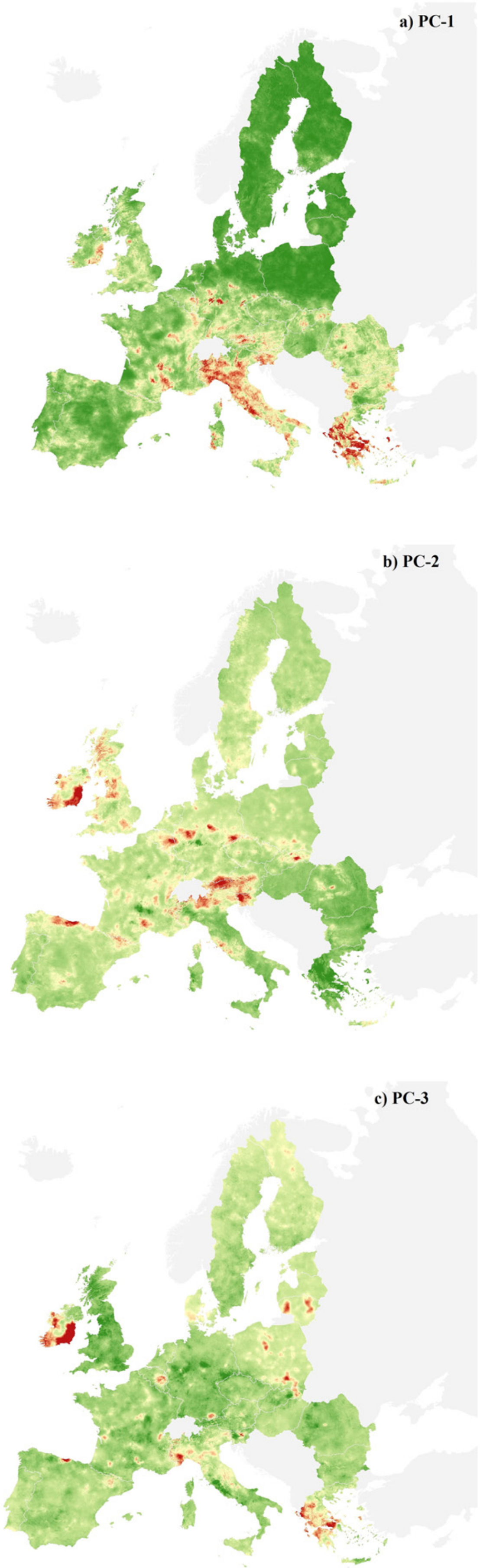

Fig. 2. Spatial representation of results of the principal component analysis on heavy metals.

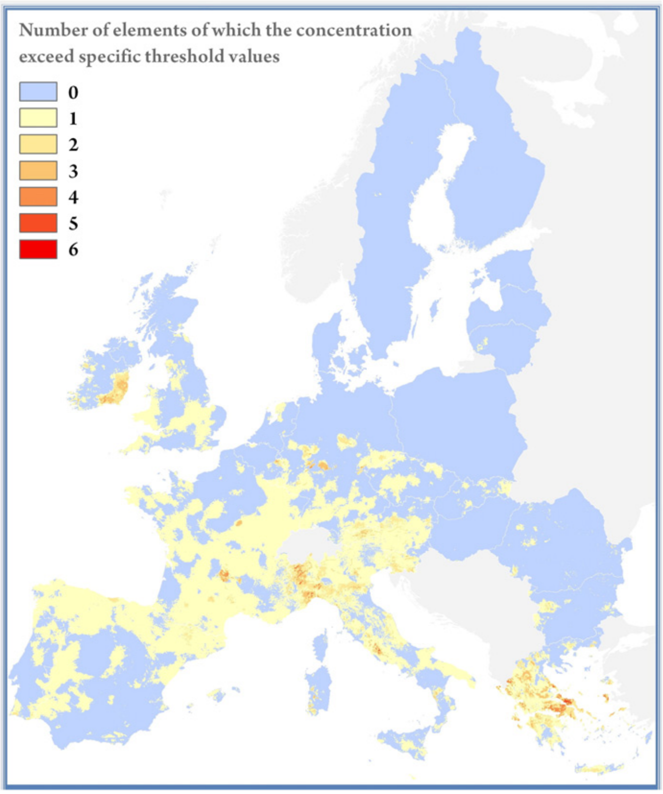

Fig. 3. Priority areas of detailed assessment of soil heavy metals.

4. Conclusion

Heavy metal contamination is among the main threats to soil-based ecosystem services, including food and feed production. Therefore a reliable information on the concentration of heavy metal in soil is essential. The LUCAS Topsoil Survey provides a unique opportunity for an appraisal of the situation of heavy metal levels in the soils of the European Union. Based on the data from the LUCAS samples we produced a series of maps which predict the concentrations of As, Cd, Cr, Cu, Pb, Zn, Sb. Co and Ni in the EU. Most of these elements remain under the corresponding threshold values in the majority of the land of the EU. However, one or more of the elements exceed the applied threshold concentration on 1.2 M km^2, which is 28.3% of the total surface area of the EU. In particular regions in Western Central Europe, Central Italy, Greece and South-East Ireland are in concern, where detailed assessment and monitoring is suggested. While natural background concentrations and anthropogenic pollution cannot be definitely separated on the basis of the maps presented, some tendencies and hotspots were identified. The main geological divide in the continent between regions with very low and higher concentration of most of the studied elements is the border of the last glaciation, running through around the 55° latitude. While isolated cases of highly polluted sites can be occasionally found in any regions of the continent, some of the larger historical and recent industrial and mining areas show elevated concentrations of As, Cd, Pb and Hg on most of their areas. These areas, together with those regions where one or more heavy metal show concentrations above the investigation thresholds are proposed to be priority areas for further detailed assessment.

Table 3
Area extent of soil cover with heavy metal concentration for further assessment.

	Area [km^2]	Area [% of the total in the EU]
No threshold exceeded (no further assessment needed)	3,087,524	71.68%
Threshold exceeded for 1 element	1,091,013	25.33%
Threshold exceeded for 2 elements	95,372	2.21%
Threshold exceeded for 3 elements	27,036	0.63%
Threshold exceeded for 4 elements	5324	0.12%
Threshold exceeded for 5 elements	880	0.02%
Threshold exceeded for 6 elements	1	<0.01%

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