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A regional approach to the environmental risk assessment - Human health risk assessment case study in the Campania region

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ABSTRACT

Environmental quality is fundamental for the wellbeing of human life. Environmental risk assessment and analysis have a crucial role in the evaluation of human health risk, especially in intensive urbanized and industrialized areas, such as the Campania region (Italy). In Italy, after the Legislative Decree 152/2006, the environmental risk assessment has become mandatory for contaminated lands such as brownfield sites.

For the purposes of the present study 3535 topsoil samples were collected across the whole regional territory. The concentrations of 53 elements have been determined by aqua regia extraction followed by a combination of ICP-MS and ICP-AES methods.

A new approach to assess/rank environmental risk was applied by using geospatial analysis in a GIS platform to adapt a European-wide accepted methodology for the preliminary assessment of human health risks at single contaminated sites to a regional scale.

The methodology chosen for the risk assessment procedures is the PRA.MS (Preliminary Risk Assessment Model for the identification and assessment of probelm areas for Soil contamination in Europe). Following the PRA.MS guidelines, a conceptual model for the human health risk assessment in the Campania region has been based on four different exposure routes: 1) dispersion of contaminants in groundwater, 2) dispersion in surface water, 3) dispersion in air, and 4) direct contact with the contaminated media (soil). The source, pathway and receptor for each exposure route are scored fusing a quantitative or qualitative analysis of some characteristic features (parameters).

A total of 14 representative parameters were chosen, based on the available regional data for Campania. Starting from the values of these parameters, the information is aggregated to higher levels in several steps, adopting a mixed additive and multiplicative algorithm, up to the overall risk score. The final risk map is classified into four risk classes. This map is useful for identifying high risk areas, where monitoring and more detailed analysis has to be carried out.

1. Introduction

In this paper a methodology for assessing the human health risk, based on spatial analysis, is presented. An existing European-wide methodology for the preliminary assessment of human health risk at single contaminated sites (PRA.MS, EEA, 2004) is adapted to evaluate the risk at the regional level, by using geospatial analysis in GIS environment. The starting point is the systematic collection of topsoil samples in the Campania region, an intensely populated and industrialized area in Southern Italy, and the determination of potentially hazardous elements.

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1.1. State of the art of soil policies

Soil is a complex mixture of mineral nutrients, organic matter, water, air, and living organisms and its function is to sustain biological productivity, maintain environmental quality, and support human health and habitations (Kalu and Anup, 2015). Being a boundary between rocks, sediments and ground-water on one side, and vegetation, air, rain, surface water, fauna and human beings on the other, it is an essential factor, acting as regulator of ground-water quality, for filtering harmful chemical elements, for biological and social activities, for agriculture and forestry productions, and at the same time the soil is subject to the effect of contaminants and waste production (Costabile et al., 2004). Soil contamination is caused mainly by atmospheric fallout from various sources, the most important being industrial and traffic emissions, but other main emission sources of potentially hazardous elements are the waste producing areas (including hospital/industrial/household/

Fig. 1. Location of Campania region.

municipal solid and liquid wastes, etc., Bou Kheir et al., 2014). Three million are the estimated potentially contaminated sites in the countries of the European Union, of which about 250,000 are actually contaminated and in need of remediation (EEA, 2007). Once the soil is degraded, it does not easily reach a new equilibrium, as the persistence and residence time of many elements and chemical compounds are two variables that should be considered in environmental risk assessment studies (Pierzynski et al., 2005). Persistence refers to the tendency of a substance to degrade in the environment, whereas residence time refers to the length of time a substance remains in an environmental compartment (e.g., soil, groundwater). For example, the mean residence times of lead is soil is between 200 and 250 years (Kabata Pendias and Pendias, 1984; Klaminder et al., 2006).

Due to the non-biodegradability of potentially hazardous elements, they can easily accumulate in the soil and enter in the food chain, having a significant effect on human health in the long term (Bou Kheir et al., 2014). Despite its importance and fragility, and the attempt to safeguard it as a significant resource by the European Commission's "Thematic Strategy on Soil" (Van-Camp et al., 2004), it is still not considered by many people as a resource to be preserved.

Starting from the 1980's several incidents attracted the major mass media attention (e.g. Love Canal, New York State; Times Beach, Missouri; Lekkerkerk, The Netherlands) and motivated politicians to understand that pollution should be removed or contained completely (Ferguson, 1999). Some countries in Europe are pioneering in assessing contaminated sites, such as the United Kingdom, Denmark, Germany, and especially The Netherlands, which developed an approach after the Lekkerkerk incident, and was used in many other countries. Other countries, such as France, Italy and Portugal, developed much later specific legislation for contaminated land and remediation (Ferguson, 1999). Of course, in this account the significant work on risk assessment of the United States Environmental Protection Agency should be mentioned (US EPA, 1989, 1991a,b, 1998a,b).

In May 1990 the European Environment Agency (EEA) was established by Council Regulation EEC 1210/90 and started its operations in Copenhagen in July 1994. The EEA's mission is to contribute to the improvement of the environment in Europe and to support sustainable development through the provision of relevant, reliable, targeted and timely information to policy-makers and the general public (EEA, 2004).

During the last years, some national environmental agencies developed in many industrialized countries ecological and human health risk assessment procedures for contaminated sites, widely implemented by legislation (APAT, 2008; DEFRA, 2011; DoE, 1995; Health Canada, 2010a, 2010b; US EPA, 1991a,b, 2005, Albanese et al., 2014).

Several Technical Working Groups (TWGs), launched in 2003 in order to support the preparation of the Soil Thematic Strategy (STS) for the protection of soils, recommended that monitoring of soil should be limited only to identified potentially risk areas, as opposed to monitoring systems covering the entire territory. Focusing monitoring in selected areas would help to define priorities, increase efficiency of monitoring activities and reduce costs (EEA, 2004). The limited financial resources in most countries, is another factor that forces to reduce as much as possible the remediation strategies. Therefore, there is a strong need to develop, so called, relative risk assessment methodologies, which aim to identify and select the potential hazardous areas to be investigated more thoroughly or in order to prioritize the remediation actions (Pizzol et al., 2011).

In 2005, the EEA published a review of 27 existing and documented international methodologies for preliminary and simplified risk assessment of (potentially) contaminated sites, already in use in member countries and overseas at national or regional level for the prioritization and planning of soil remediation and protection programmes. All the reviewed methodologies adopt a qualitative (or semi-quantitative) approach to the assessment of site risks, describing risks in term of scores, rather than absolute estimates of health/ecological impacts (EEA, 2004). The scoring system is based on several parameters. The identification and listing of common parameters, used in the reviewed methodologies and their harmonization, helped in the selection of a set of parameters to be included in the proposal for a methodology for the identification of potentially hazardous areas for soil contamination in Europe. This methodology, called PRA.MS (Preliminary Risk Assessment Model for the identification and assessment of problem areas for Soil contamination in Europe; EEA, 2005a,b, 2006) is a tiered approach, that starts from a preliminary selection of the sites, which are potentially hazardous, based on a source size criteria, and implementing it with fundamental risk elements, such as source hazard, pathways and receptor information (EEA, 2005a). Potentially hazardous areas are defined in the PRA.MS as areas where soil contamination is considered to pose significant risks to human health and/or ecosystems with impacts beyond the local environment (Quercia et al., 2006). The main sources of contamination are municipal and industrial waste disposals, mining sites, and industries.

1.2. Purpose of the research

The only drawback that the PRA.MS and the other 27 reviewed methodologies have is the absence of spatial analysis (Pizzol et al., 2011). The traditional way to evaluate the human health risk is by calculating the product of the Hazard, generated by the presence in the environment of one or more contaminating agents, and the Vulnerability, which includes the possibility for a receptor (humans) to come in contact with the potentially toxic substance released by the contaminating agent through an exposure pathway. The evaluation of vulnerability involves the identification of the factors affecting the receptor's vulnerability (Zabeo et al., 2011) for each of the identified exposure route. Due to the heterogeneity of many variables involved in this process, the human health risk assessment is used at the scale of a site (Tristán

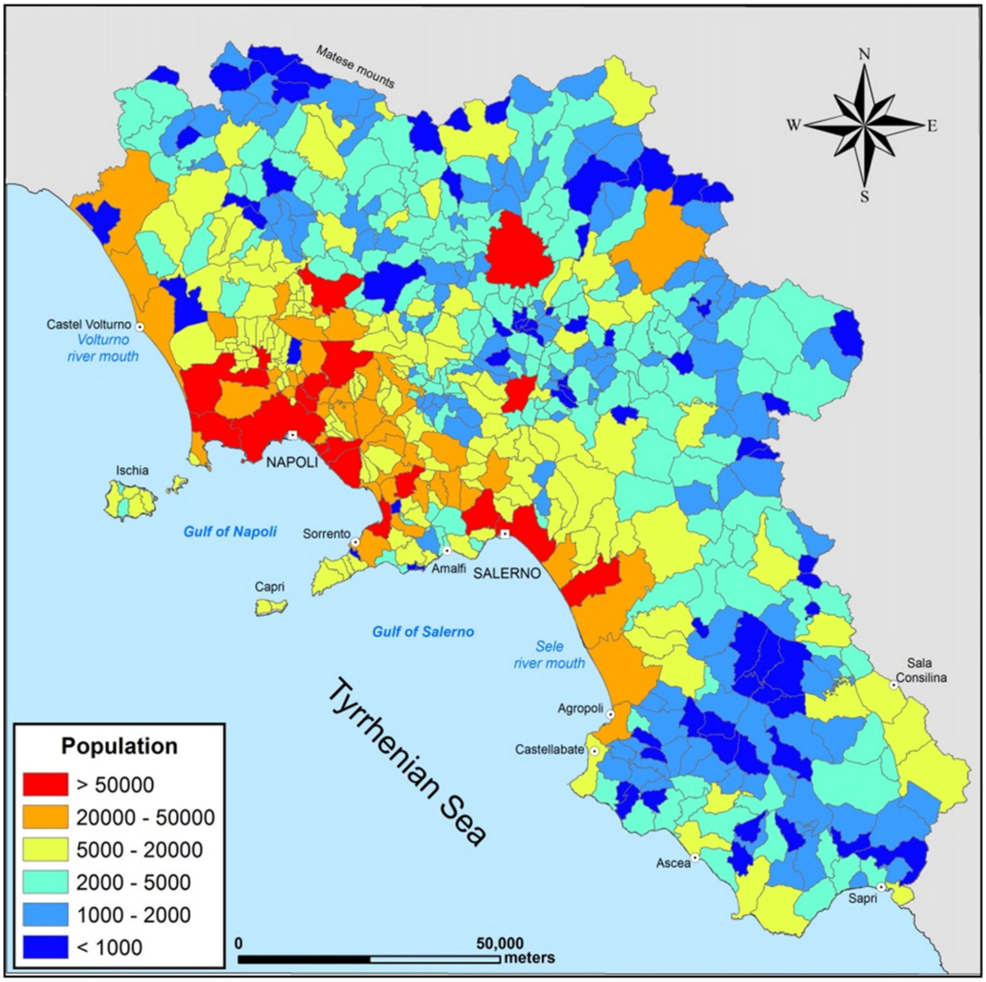

Fig. 2. Municipality based distribution of population density in Campania region.

et al., 2000; Demetriades, 2011; Albanese et al., 2014), while the regional evaluation is very difficult.

The objective of this paper is to implement the PRA.MS methodology, integrating the model of relative risk assessment for single contaminated sites, with spatial analysis procedures, developing a regional risk assessment methodology, which can be used by the regional administrations to select at regional level the potentially hazardous areas and to prioritize them. In order to support the spatial assessment of contaminated sites at the regional scale, as mentioned above, the most suitable tool is the Geographic Information Systems (GIS) (Pizzol et al., 2011). The GIS is the main tool to manage, manipulate, process, analyze, map, and spatially organize the data in order to facilitate the vulnerability analysis (Zabeo et al., 2011). Despite this, there are very few applications in risk or exposure assessment (Tristán et al., 2000 and references therein).

For the purpose of this work 3535 topsoil samples were used, which were collected over the whole Campanian territory, and already statistically processed by Buccianti et al. (2015) using Compositional Data Analysis (CoDA). In Buccianti et al. (2015) the multi-element data archive describing the topsoil geochemistry of Campania region has been used to propose a new compositional data methodology, with which to characterize the structure of the data and identify background/baseline composition.

The identification of potentially hazardous areas is necessary for developing an efficient monitoring system and to produce a ranking of areas at risk, which will be used as a reference for the development of intervention plans, and for better addressing the utilization of available resources to environmental remediation of widely contaminated regions.

2. Study area

Campania region is situated in south Italy (Fig. 1), and it lies between the Tyrrhenian Sea to the West and the Apennine mountain chain to the East. The whole territory covers an area of about 13,660 km^2. The region comprises five provinces: Napoli, Salerno, Caserta, Avellino and Benevento, in order of population density (Fig. 2). The Campania region is the first most densely populated in Italy with 429 inhabitants/km^2, and the third in the number of inhabitants (ISTAT, 2015), with >50% concentrated in the Naples metropolitan area (Albanese et al., 2007). Some inner mountainous areas (Mt. Matese, Cilento area, etc.) are characterized by the presence of small cities with a very low population density, traditionally dedicated to agricultural and pastoral activities.

2.1. Geological and geomorphological settings

The geology of Campania region is the result of different processes. It can generally be distinguished in the eastern hilly/mountainous area and the western coastal low lying part, occupied by alluvial plains. The eastern area is made up of the southern Apennine mountain chain, oriented NE-SW, resulting both from compressive tectonic events, related to the subduction followed by the roll-back of the Adria plate, and from the extensional tectonics associated with the late Miocene opening of the Tyrrhenian sea (Bonardi et al., 2009). Due to the extensional forces,

Fig. 3. Land Cover distribution for Campania region (Corine Land Cover, 2012).

the western area of the Campania region is occupied by a large subsident graben (from Pliocene-Pleistocene up to 5 km, Ippolito et al., 1973), filled with sediments originating from the erosion of the Apennine ridge and from products of intensive volcanic activity. The whole area of the graben presently constitutes the Campanian and Sele plains. The volcanism in the Campanian plain is part of the Roman Co-magmatic Province (Washington, 1906), getting younger from Tuscany to Campania. The conditions were ideal for triggering fissure volcanic activities (Peccerillo, 2005 and references therein), with different ignimbrite eruptions (Campania Ignimbrites), as well as volcanic complex formations (Roccamonfina, in the north-western sector of the region; Campi Flegrei and Ischia, along the western border of the region; Mt. Somma-Vesuvio) (De Vivo, 2006a and references therein; De Vivo et al., 2001; De Vivo et al., 2010; Rolandi et al., 2003 and references therein; Milia and Torrente, 2000; Milia and Torrente, 2013). The volcanic rocks (lava and pyroclastics), dated from about 600 ka to present, are represented by potassic to ultrapotassic rocks (De Vivo et al., 2001; Rolandi et al., 2003), with alkaline magmatism, characterized by high K_2O content of (De Vivo et al., 2010; Peccerillo, 2005). The main sedimentary lithologies consist of: (i) limestone, dolostone, siliceous schist and terrigenous sediments (clays, siltstone, sandstone, conglomerate), part of the Mesozoic Units, which characterize mostly the external Apennine domains, (ii) the Neogene units, made up mostly of silico-clastic, carbonatic and evaporitic sediments, and (iii) Quaternary formations represented by alluvial, lacustrine and evaporitic sediments and by pyroclastic fall and flow deposits, occurring mainly in the Campania plain.

2.2. Climate and hydrogeology

The Campania region has a Mediterranean climate, with hot dry summers and moderate cool rainy winters (Ducci and Tranfaglia 2005). The differences in climate between the high mountains in the interior and the coastal areas are quite large. The maximum temperature during the winter times (January) varies between 11 and 13 °C along the coast, and 5–8 °C on the mountainous interior zones, and between 28 and 31 °C/25–28 °C during summer; the minimum temperature rarely is below 5–6 °C along the coast, while in the interior can be very low during severe winters.

The rainfall regime in Campania has a maximum in autumn/winter, mainly influenced by mountain chains, in terms elevation, location of ridges (barrier effect) and proximity to Tyrrhenian Sea (Ducci and Tranfaglia, 2005). The maximum mean annual rainfall (>1100 mm/yr) occurs in the central part of the Apennine ridge, especially in the Partenio and Picentini Regional Parks, and in the southern Cilento area (up to 1800–2000 mm/yr). The lowest precipitation (600–700 mm/yr) occurs along the coastal plains and in the northeastern part, on the other side of the Appenninc watershed, with the Benevento province (Ducci and Tranfaglia, 2005; Mazzarella and Fortelli, 2012).

The hydrography of Campania region is quite simple with two main rivers: Volturno River is 170 km long, with a hydrographic basin covering 5600 km^2, covering about 40% of the regional territory, located in the northern part of the region; the second river is the Sele, 65 km long and extending over an area of 3200 km^2 in the southern part of the region.

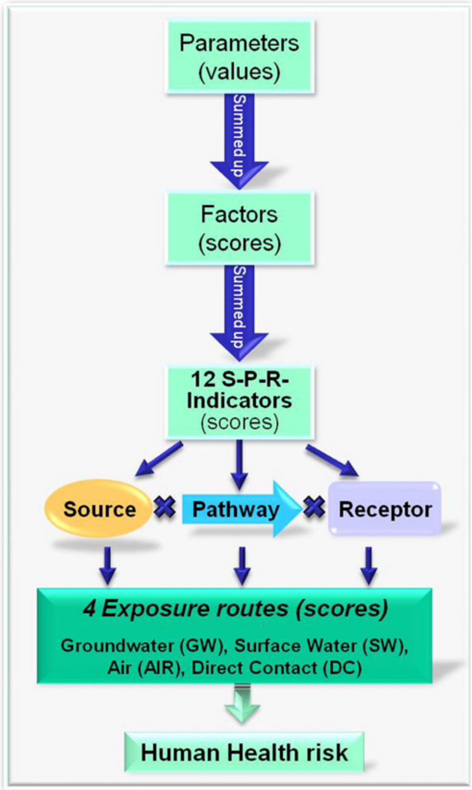

Fig. 4. General framework of the PRA.MS methodology (modified after Quercia et al., 2006).

2.3. Economy

The economy of Campania region exhibits from the 1990's a distinct change, transforming the main specializations from the traditional industrial/productive activities, such as mechanical or textile industries, to small businesses, centered on services and tourism. Commercial activities have a 35% share of the economy, services 25%, constructions 10%; industries and restaurants/accommodations have an equal share at 9%, followed by transport and warehousing (7%), health care centers and assistance (5%), other services (4%) and instruction (1%) (Banca d'Italia, 2015).

As it can be observed from the Land Use Cover map (Fig. 3), agriculture is the main activity in the Campania region. In the North, the agricultural activities cover >50% of the available land, while in the South farming is mostly developed along the coast, due to the occurrence of mountainous areas. The pyroclastic deposits, covering most of the carbonates and flood-plain deposits located along the coast, help the productivity of this territory. Despite this natural fertility of the volcanic soil, rich in "mineral nutrients", Campania is one of the Italian regions with the highest consumption of fertilizers, containing Cd, Cu and V as contaminants.

After Sardinia and Calabria Regions, Campania is the Southern region with the largest forest area and with the highest percentage (30%) of land covered with national and regional parks and nature reserves (Lombardo and Ansanelli, 2011).

Industries are mostly distributed in the northern half of the regional territory. The main industrial activities are connected to vegetable canning, textile, clothes and tanning production. Industrial related contamination can be ascribed to lack of maintenance of the purification and waste-water systems that can lead to a severe contamination of stream water, ground-water and sediment.

No economic mineral deposits occur in Campania; only a few minor bauxite mineral occurrences–of no economic relevance – are located in the Mesozoic rocks of Mt. Matese in the Apennine Mountains (Albanese et al., 2007).

3. Materials and methods

3.1. Sampling and sample preparation

To reach the aim of the reported research, during 2013 and 2014, a total of 3535 topsoil samples were collected over the whole Campanian territory with a nominal density of 1 sample/4 km^2.

The sampling procedures followed the international guidelines established by the FOREGS Geochemistry Group (Salminen et al., 1998): about 1.5 kg soil was collected from a depth between 5 and 15 cm after the removal of the vegetation cover. For each sample field notes were recorded, namely the alphanumerical code, the spatial coordinates of the sampling site and any other useful information about the sampling site, such as local geology, topography, type of soil and vegetation, land use and indication of potential contaminating anthropogenic activities (roads, industries, use of pesticides, insecticides etc.). Duplicate field samples were collected at every 20th sampling site, in order to estimate the sampling variability.

The samples were dried under infra-red lamps at a temperature below 35 °C, and then disaggregated in a ceramic mortar and sieved through a 2 mm nylon mesh. Following homogenization of the <2 mm grain size fraction, at least 30 g of each routine and field duplicate sample was placed in two small plastic bags for laboratory analysis, and the remaining amount was stored for future reference.

3.2. Laboratory analysis

The analysis were conducted in the Bureau Veritas Analytical Laboratories Ltd. (Vancouver, Canada), where they determined by a combination of ICP-MS and ICP-AES, following a hot aqua regia digestion, the concentrations of 53 elements: Ag, Al, As, Au, B, Ba, Be, Bi, Ca, Cd, Ce, Co, Cr, Cs, Cu, Fe, Ga, Ge, Hf, Hg, In, K, La, Li, Mg, Mn, Mo, Na, Nb, Ni, P, Pb, Pd, Pt, Rb, Re, S, Sb, Sc, Se, Sn, Sr, Ta, Te, Th, Ti, Tl, U, V,W, Y, Zn and Zr.

Precision of the analytical results was calculated using the data of 29 in-house replicates, and 5 blind project duplicates (median value of the Relative Percentage Difference, RPD = 1.3%). Accuracy was determined using in-house reference materials (STD DS9, STD DS10, STD DS11,

Table 1
Parameters chosen from the PRA.MS methodology for the human health risk assessment.

Groundwater (GW)	Surface water (SW)	Air (AIR)	Direct contact (DC)	
Potential hazard map	Potential hazard map	Potential hazard map	Potential hazard map	Source
Lithology of unsaturated zone	Slope	Particulate mobility		Pathway
Infiltration	Flooding risk			
	Surface water flow rate			
Distance of site to drinking water supply	Surface water use	Land use at site	Land use at site	Receptor
	Minimum distance from surface body	Distance to the nearest residential area	Distance to the nearest residential area	

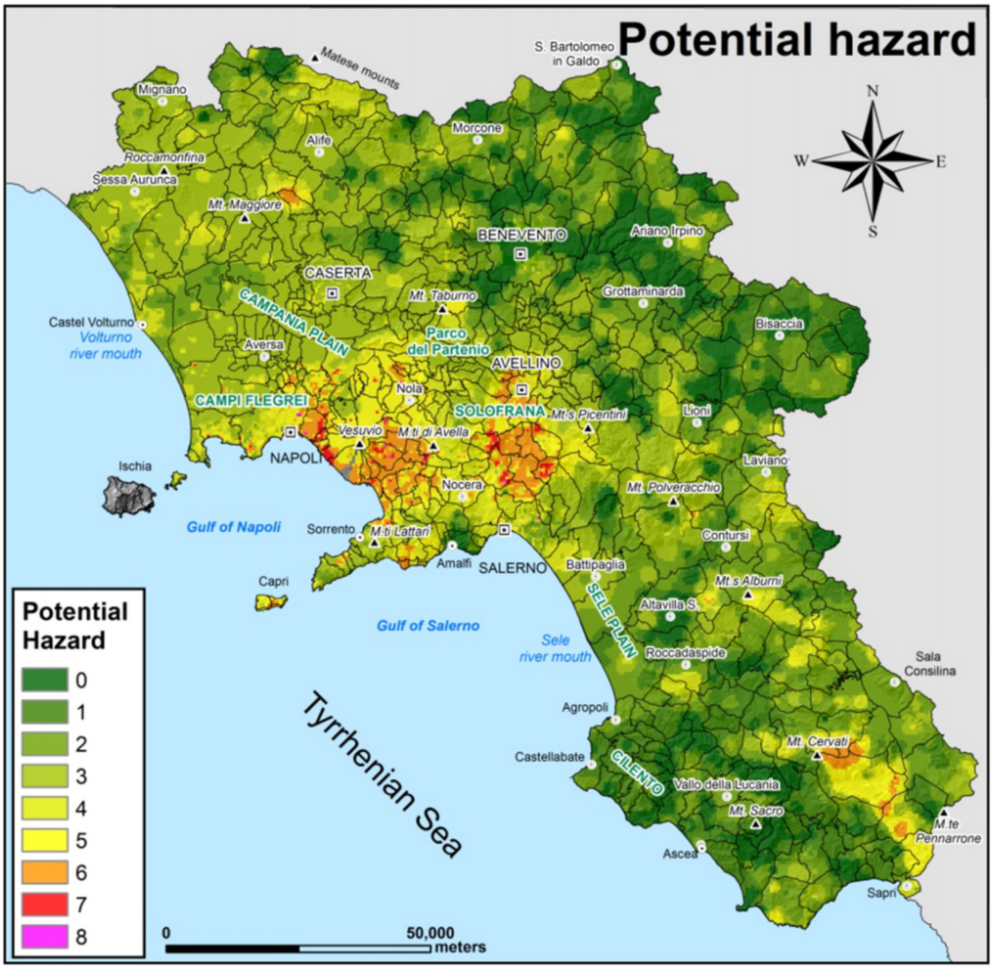

Fig. 5. Composite additive map of the Hazard due to the presence of potentially toxic elements (Sb, As, Be, Cd, Co, Cr, Cu, Hg, Ni, Pb, Se, Sn, Tl, V, Zn) in the topsoil of Campania region.

STDOXC109, median value 2.2%). Method detection limits for Al, Ca, Fe, K, Mg, Na, P, S and Ti range from 0.001 wt.% (Na, P, Ti) to 0.02 wt.% (S) and from 0.01 mg/kg to 5 mg/kg for all the other elements (Buccianti et al., 2015).

4. Risk assessment methods

4.1. Risk assessment using PRA.MS methodology

The general framework for the risk assessment adopted by the PRA.MS methodology (EEA, 2005b) is shown in Fig. 4 and it consists of several steps:

1) The exposure routes relevant for the human health risk are the following: Groundwater (GW), Surface Water (SW), Air Particulates (AIR), and Direct Contact (DC);
2) For each exposure route, a Source-Pathway-Receptor model is assumed to assess the human health risk;
3) Representative parameters of the Source, Pathway and Receptor for the different exposure routes, are established by taking the common parameters of the existing methodologies and by their harmonization;
4) Parameters, alone or aggregated, represents factors;
5) The factors are given scores based on the qualitative or quantitative value of the parameters and have different weights depending on how well they describe the exposure routes indicators;
6) Factor scores are added up in order to aggregate each Source-Pathway-Receptor Indicator (as indicated in Fig. 4, there are 12 S-P-R Indicators);
7) S-P-R Indicators are multiplied for each of the exposure routes, to obtain the corresponding scores of the four exposure routes, and;
8) The overall risk is given a score by computing the root mean square of all exposure routes scores.

The PRA.MS methodology is a tiered approach, which starts from a pre-selection of the potentially contaminated sites (Tier 0), and passes through two Tiers (1 and 2). The choice of which Tier is better to use depends on the quality and availability of the required data. In this study it was decided to use Tier 2, which is the more detailed approach for the human health risk assessment and is based on quantitative data. A fundamental step for the work has been the collection of the georeferenced environmental regional data, in a shapefile format, for the Campania region. On the basis of the available data, the representative parameters of the Source-Pathway-Receptor for the different exposure routes were selected, and are tabulated in Table 1. The scores were normalized to 100, even if not all the parameters were available for a certain Source-Pathway-Receptor Indicator.

4.2. The sources

4.2.1. The hazard map

The first measure that is required in the Tier 2 of the PRA.MS methodology for featuring the Source is the chemical characterization of the soil contamination: first are recognized the chemical

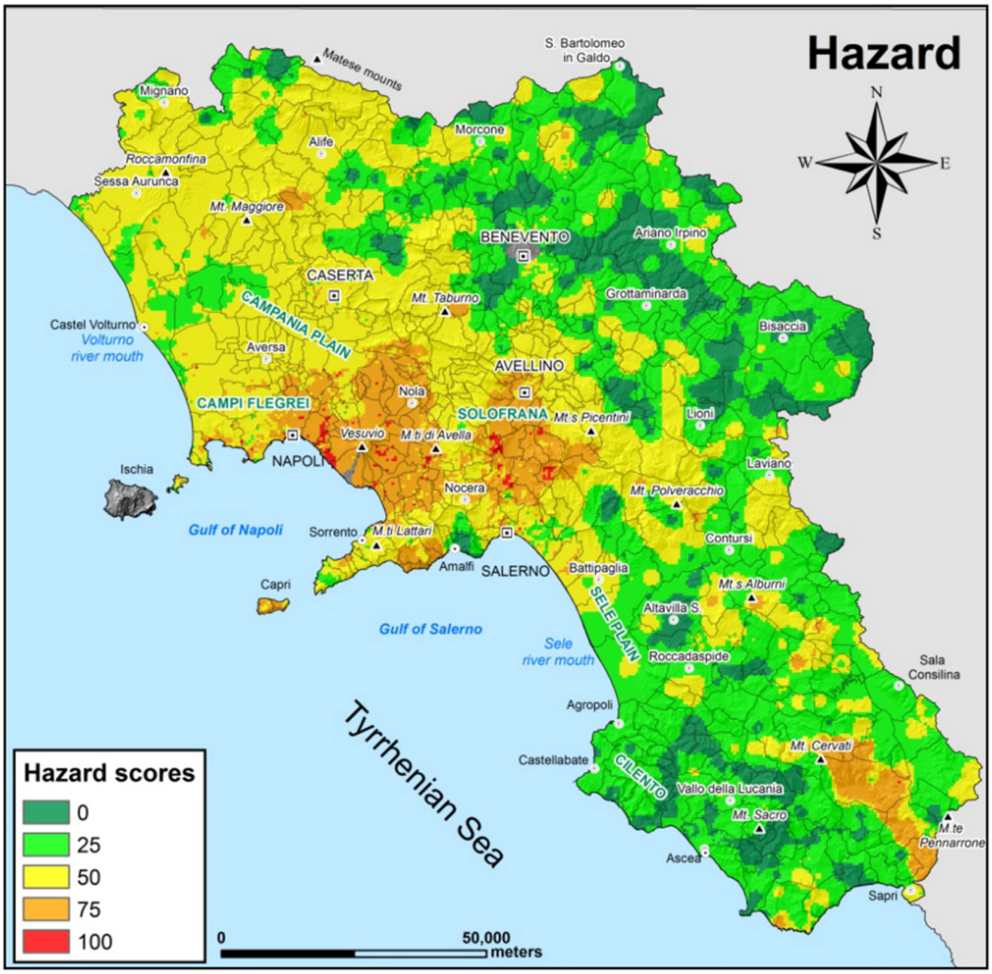

Fig. 6. Reclassification of the hazard map of Campania region according to the scores in Table 2.

elements at each site, then their chemical toxicity and scores are evaluated. The PRA.MS model is not implemented for cases where more than one contaminant is present at the same site. Hence, in the PRA.MS methodology, only the most toxic contaminant, for each exposure route is taken into account.

In the case of a regional risk assessment, the risk at single industrial or mining contaminated sites is not evaluated, but the assessment is extended to cover a wider territory. Using the GIS tool, is it possible to treat each pixel belonging to the regional territory as a potential source. Starting from the chemical characterization of the topsoil samples, covering the entire Campania region (Buccianti et al., 2015), described above, the hazard map was plotted, with a pixel dimension of 20 m, which contains all the information about the presence, the amount and the toxicity of multiple contaminants.

The determination of the concentration of 53 elements for each sample was useful to elaborate a detailed statistical and cartographical analysis of the Campania region (Buccianti et al., 2015, De Vivo et al., 2016), and is a reference guide for all further elaborations. For the purpose of this study, only the 15 potentially toxic elements (Sb, As, Be, Cd, Co, Cr, Cu, Hg, Ni, Pb, Se, Sn, Tl, V, Zn) for which the Italian legislation (D.Lgs. 152/06) established trigger and action limits were taken into account, by defining the contamination threshold values (CSC) for the amount of chemical elements in topsoil or water samples. The D.Lgs. 152/06 promotes a requalification of human life by an improvement of the environmental quality. The law assumes that, if a contaminant concentration in a soil exceeds the CSC value, a follow-up risk assessment must be carried out at the site. Once a follow-up risk assessment has been carried out, the remediation can be applied only if the contaminant concentration exceeds the risk threshold values (CSR), valid only in the specific site of interest. Two are the CSC values indicated by the legislation, one regarding the hazard in residential sites, and one with a higher value for industrial and commercial sites. Because the CSC values are established for the human health risk, they can used directly for the definition of the hazard; in particular, the conservative CSC values for residential sites are considered.

The interpolated grids of the spatial distribution of these elements, obtained by multifractal IDW method using the dedicated GIS GeoDAS™ geochemistry software (Cheng, 2003), were the basis for plotting the hazard map. To each pixel of the interpolated maps, a value of 1 was assigned if the element concentration was above the CSC, and 0 if the element concentration was below the CSC. The produced Boolean surfaces clearly show, based on the current legislation, if contamination is present, and consequently a potential hazard for the population. The soil threshold values for the

Table 2
Assigned scores for the hazard map, characterizing the source for all exposure routes. Out of the 15 potentially hazardous elements, only 8 (Zn, Tl, V, Pb, Cu, Cr, Be, As) occur in the Solofrana area; Be, Cd, As, Co, Sn, V, Tl in the southern Cilento area.

Number of potentially hazardous elements	Hazard scores
0	0
1–2	25
2–4	50
4–6	75
6–8	100

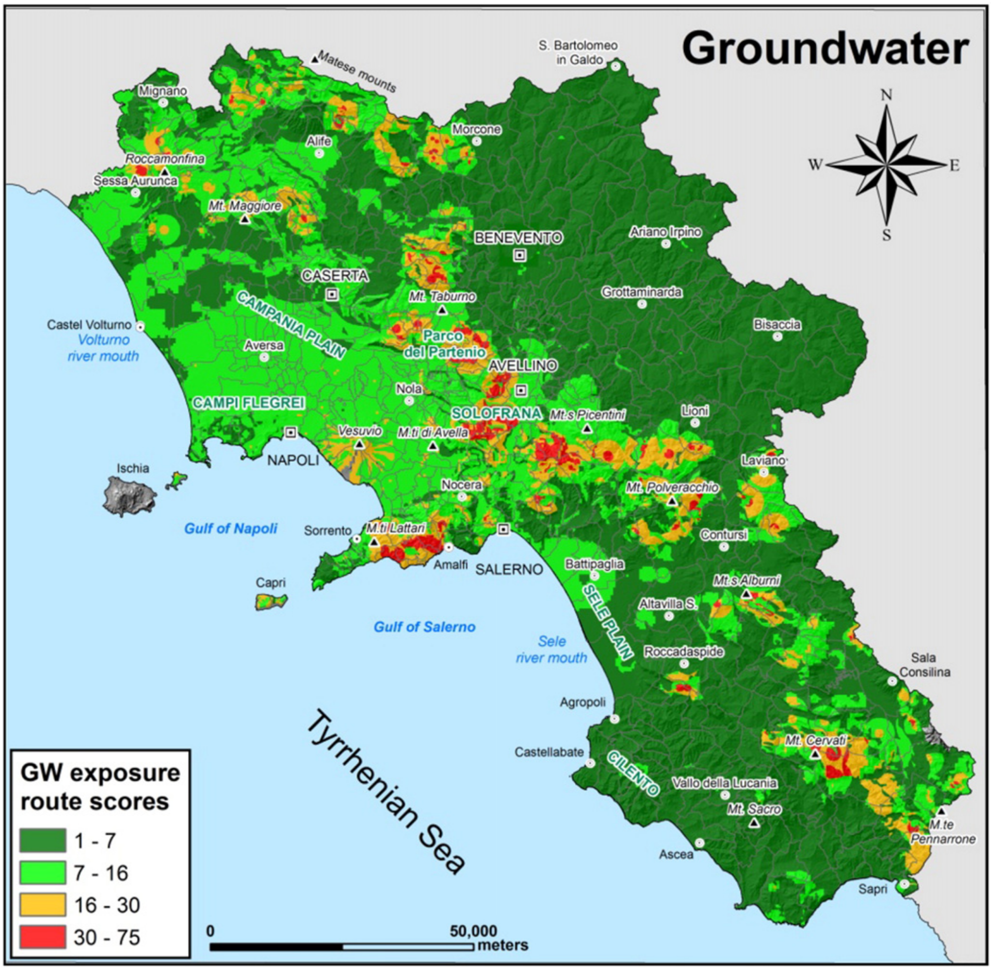

Fig. 7. Groundwater (GW) exposure route map for Campania region.

toxic elements, established by the Italian legislation, are for the potential human health risk that may be caused by contamination, and does not take into account the natural geochemical background values related to the different lithologies. In the GIS environment, by means of the Spatial Analysis module, the Boolean surfaces were summed up to obtain a composite additive map of the Hazard (Fig. 5). On the map, the higher values correspond to the areas where there is a contemporary presence of a higher number of potential toxic elements that exceed the threshold values, representing a more intense soil degradation. The map can be used as a Source Hazard characterization for further analysis. The map has been reclassified assigning four scores between 0 and 100 (Fig. 6, Table 2).

Table 3a
Scoring model for the groundwater pathway indicator; lithology factor scores evaluated through the permeability of the hydrogeological complexes.

Permeability of hydrogeological complexes	Lithology factor scores
High permeability – fractured igneous rocks and karst limestone	78
Moderate permeability – moderately permeable dolomite, cemented sandstone, molasse, pyroclastic fall deposits	56
Low permeability – clayey silt, less permeable limestone, turbidite	33
Very low permeability – clayey limestone, prevalentely clay	11

4.3. Exposure routes

4.3.1. Groundwater (GW) map

The GW exposure route map (Fig. 7) is obtained by means of spatial analysis in ArcGIS software, using the following expression:

$$GW_{er} = (S_{gw} * P_{gw} * R_{gw}) \div 10^4$$

where: S_{gw} is the groundwater hazard map, P_{gw} is the GW Pathway Indicator, and R_{gw} is the GW Receptor Indicator.

The classification of the four exposure route maps is computed with the standard Natural Breaks in the ArcGIS software, assigning four intervals.

Table 3b
Scoring model for the groundwater pathway indicator; infiltration factor scores evaluated through the mean annual precipitation values (mm/yr).

Mean annual precipitation (mm/yr)	Infiltration factor scores
<1100	22
900–1100	18
700–900	13
500–700	9
300–500	4
<300	0

Table 4
Scoring model for the Groundwater Receptor Indicator, by means of the distance of the Source from the nearest drinking water supply.

Distance from the drinking water supply (m)	Distance factor scores
0–150	100
150–400	86
400–900	71
900–1500	57
1500–3000	43
>3000	29

Table 5a
Scoring model for the Surface Water Pathway Indicator; slope factor scores.

Slope	Slope factor scores
>8%	44
5%–8%	26
2%–5%	18
<2%	9

The GW Pathway Indicator is represented by the sum of two factors: (a) the lithology of the unsaturated zone and (b) the infiltration rate. The main factor that contributes to the transport of contaminants to the aquifer is the properties of the unsaturated zone that contaminants have to pass through before they reach the aquifer. The weight of this factor, which has the capacity to attenuate the dispersion of contaminants in the groundwater, is 78, while the infiltration rate has a weight of 22. As already explained, the weight of a factor is given the maximum allowed score of that factor in the PRA.MS methodology, normalized to 100. In the PRA.MS methodology the scores assigned to the factor of lithology of the unsaturated zone are grouped into 4 classes, taking into account the top layer geology, structural and hydraulic properties. In particular it evaluates the vertical hydraulic conductivity. Then other factors are evaluated in order to estimate potential release to groundwater, such as the presence and thickness of any impermeable layer, the aquifer depth from ground surface and the contaminant mobility. All these parameters cannot be evaluated at regional level, due to their variability. Moreover the vertical hydraulic conductivity is not related to the unsaturated zone, because it is usually measured under saturated flow conditions, so it should not be used in connection with a classification relating to vertical flow rates through the unsaturated zone (Lewis et al., 2006).

For scoring the lithology of the unsaturated zone the hydrogeological map of Campania region (SIT GEOPortale, 2008) was used. Before applying permeability values to the unsaturated zone (Lewis et al., 2006), this map was reclassified into four classes of scores (Table 3a), using the type of hydrogeological complexes of Campania region and their permeability. The definition of hydrogeological complex is based on the relative permeability, including similar lithologies,

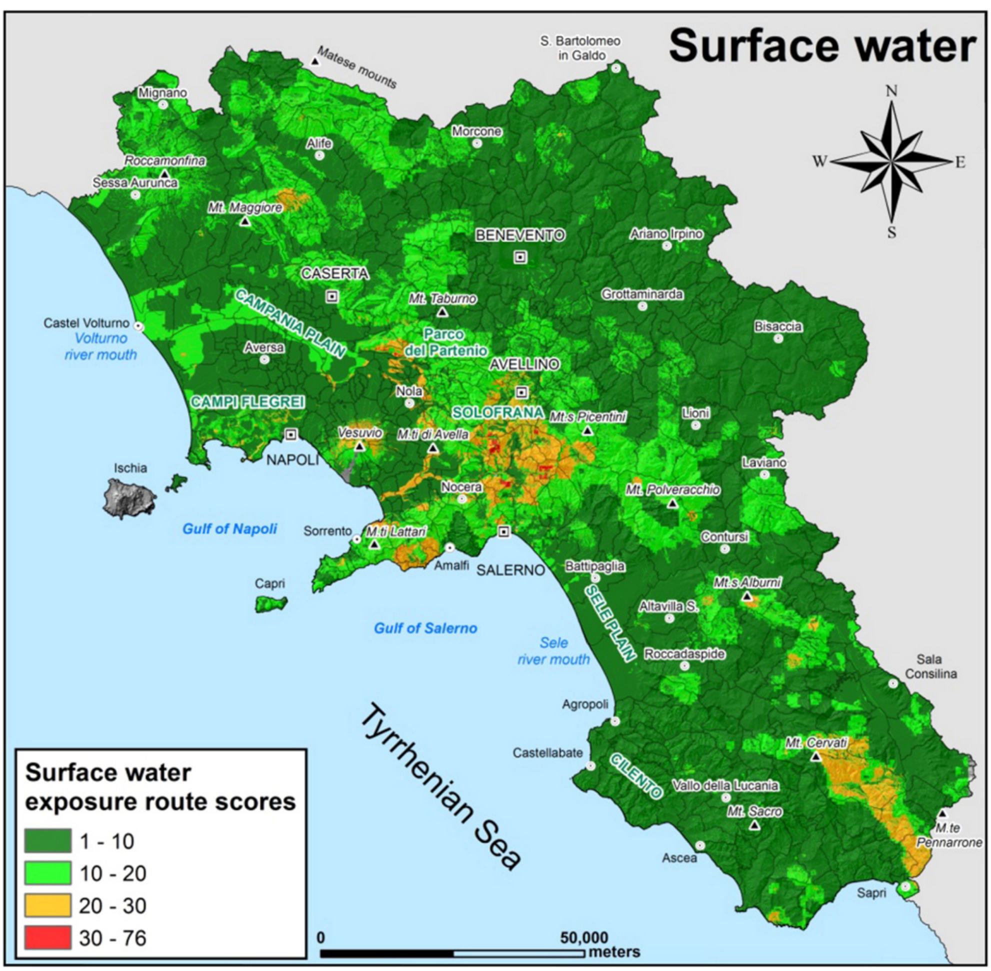

Fig. 8. Surface water (SW) exposure route map for Campania region.

Table 5b
Scoring model for the Surface Water Pathway Indicator; surface water flow rate factor scores by analyzing the stream orders.

Surface water body type – stream order	Surface water flow rate factor scores
Order 1 – small to moderate stream	25
Order 2 – moderate to large stream	20
Order 3 – large stream to river	15

with similar type of permeability and with a restricted range of relative permeability. Hence, using the hydrogeological complexes, parameters such as granulometry, fracturing, karstification and the mean annual groundwater discharge are already taken into account. An additional layer that could be evaluated is the permeability of soil (Lewis et al., 2006), especially in the cases where the soil is different from the parent material (e.g. the pyroclastic deposits on carbonate rocks in Campania region), but the intention is to modify as least the original methodology. The permeability of the lithologies of Campania region varies from: medium-high for porosity in the coastal Campanian and Sele plains; very high for the carbonates that form the Apenninic belts, with high effective infiltration due to fractures and karst; low and very low in the north-eastern part of the region, caused by the presence of silt and clay deposits.

The infiltration classification and scoring in the PRA.MS methodology are based on mean annual precipitation data. The 28 meteorological stations of the Regional Agrometeorological Network (C.A.R, 2012), covering the whole regional territory, were used (i.e. measurements of precipitation, temperature, humidity, wind velocity and surface soil humidity). In a first attempt, only the precipitation data of the last measured year (considering the total mm of rainfall during 2012 in the different stations) were used. After that, the mean annual value of the precipitation between 1999 and 2012 was calculated. For the compilation, the data relative to annual precipitation, where one or more months were missing, the data were deleted. Only the stations with >50% of the data were considered (i.e. more or equal to 7 years of measurements). Afterwards both the mean and the median values of the data were calculated, and it was found that the mean percentage difference between the mean and the median is about 5%, and only in three cases the difference is significant (20%). The interpolated map of these data was plotted using the standard IDW method.

Looking at the interpolated maps of both mean and median, it can be assumed that the mean values are effectively the most relevant to reality. The measured precipitation has an extremely high spatial variability. Hence, it is important that the interpolation does not smooth out the local variability of the data. The interpolated map was reclassified using the scores tabulated in Table 3b. The precipitation in the Campania region is not <700 mm/yr. No appreciable differences in the resulting maps of groundwater exposure route and overall risk, among the 2012 precipitation data and the mean annual values between 1999 and 2012, were observed.

The GW Receptor Indicator is represented by the distance of the nearest drinking water supply from the source. The available data about the drinking water supply in Campania come from the ISPRA-MAIS database. The map of the distance is computed by means of the Euclidean Distance tool in ArcGIS software and is reclassified according to the scores of Table 4.

The maximum scores for the GW Pathway Indicator (80–100) are located along the Apennine belt, which correspond to the zone with the highest permeability of the unsaturated zone and with the highest mean annual precipitation. Between 60 and 80 are the scores in the Campanian plain along the coast, where the precipitation is lower, but the permeability of the unsaturated zone is still quite high, with a score of 56.

The final GW exposure route map (Fig. 7) is affected by the location of the drinking water supplies on the carbonate rocks, so that the maximum scores for this exposure route are located nearby the drinking water supply wells (between 16 and 75). Scores between 7 and 16 are found on the remaining Apennine belt and on the Campanian plain, while the remaining territory, with low permeability, low precipitation and no drinking water supply, have scores between 1 and 7.

Table 6
Scoring model for the Surface Water Receptor Indicator, by means of the distance of the Source from the surface water bodies. To these scores a value of 46 was added (see text).

Distance from the surface water bodies (m)	Distance factor scores
<200	30
200–850	24
850–1700	18
1700–2600	12
2600–3600	6
>3600	0

4.3.2. Surface water (SW) map

The SW exposure route map (Fig. 8) is obtained by means of spatial analysis in ArcGIS software, through the following expression:

$$SW_{er} = (S_{sw} * P_{sw} * R_{sw}) \div 10^4$$

where: S_{sw} is the surface water hazard map, P_{sw} is the SW Pathway Indicator, and R_{sw} is the SW Receptor Indicator.

The SW Pathway Indicator, explaining the potential of contaminants to be dispersed in the surface water, depends on three factors: the terrain average slope, the flooding risk and the surface water flow rate.

For the average slope map the Digital Terrain Model 20 m for the Campania region was used. The grid was reclassified into four classes of slope (Table 5a), with the highest scores corresponding to the highest slopes, which have a large amount of run-off that contribute to the migration of contaminants in surface water. The floodplain map of the Campania region, available from the SIT database (SIT GEOPortale, 2008), did not contain the information about the flooding return periods, and this is due to the extreme heterogeneity of the data elaborated from the different Basin Authorities. Hence, the reason for only producing a Boolean surface, assigning the maximum allowed score for the flooding risk (31) indiscriminately to all the floodplain areas, and a score of zero for the remaining territory. The surface water flow rate map was obtained from the surface water map of Campania region, available from the project DBPrior10K (CISIS, 2007). The surface water body types were differentiated with respect to stream size and stream order (Table 5b). The scores are higher for low stream orders, because there is a general decreasing trend of contaminant concentrations with increasing stream order, explained by a dilution effect (Kang et al., 2008).

The SW Receptor Indicator is the minimum distance of the surface water body from the source. The Euclidean Distance map was developed from the same surface water map of Campania region used previously, and then reclassified to 6 classes of scores (Table 6). To these scores, the value of 46 was added; it is assumed, in fact, that the most common surface water use in Campania region is the irrigation with food crop, having the score of 46 in the PRA.MS methodology.

In the final SW exposure route map (Fig. 8) two are the main areas with higher vulnerability of the surface water bodies; the first is between the Avellino and Salerno provinces, and the second is in the south eastern part of the Cilento area.

4.3.3. Air Particulate map

The Air Particulate exposure route map (Fig. 9) is obtained by means of the spatial analysis module in ArcGIS software, through the following expression:

$$Air_{er} = (S_{air} * P_{air} * R_{air}) \div 10^4$$

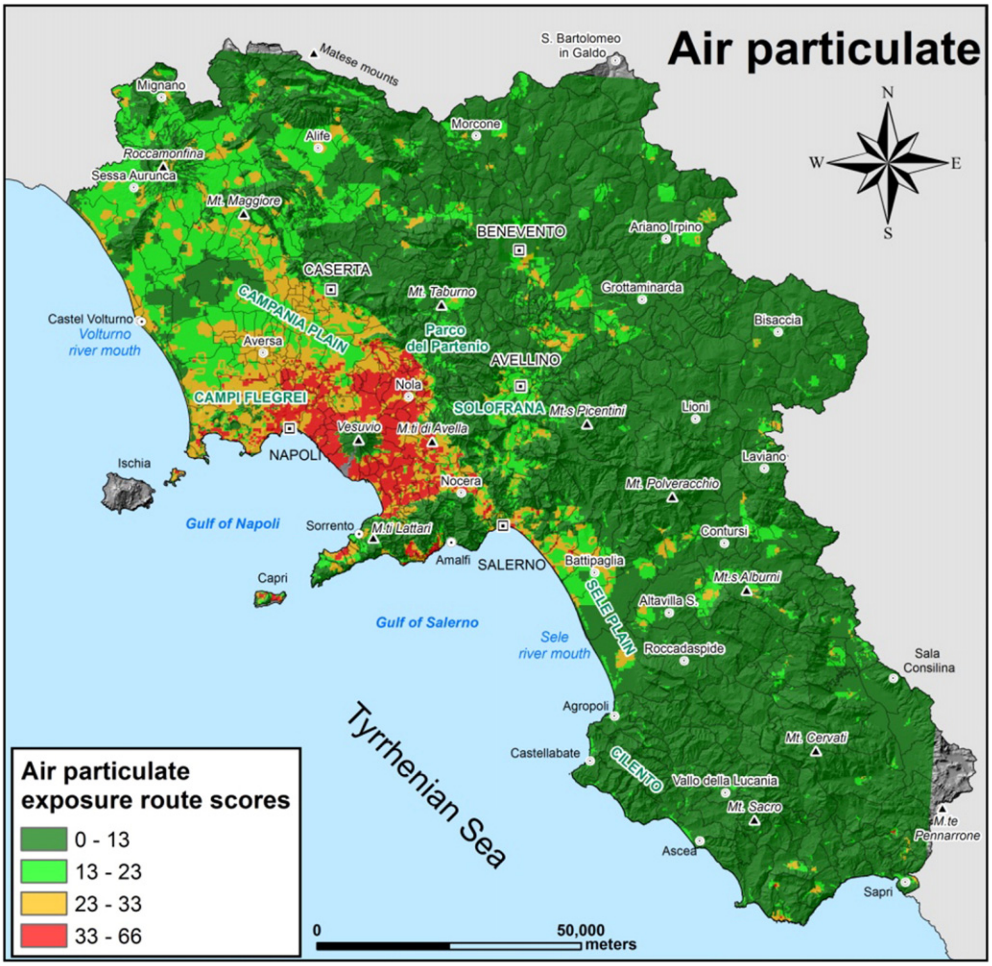

Fig. 9. Air Particulate exposure route map for Campania region.

where: S_{air} is the Air Particulate hazard map, P_{air} is the Air Particulate Pathway Indicator, and R_{air} is the Air Particulate Receptor Indicator.

The only parameter that characterizes the Air Particulate Pathway Indicator is the Particulate Mobility, related to the possibility for the soil solid particles to be transported away from the contaminated source. The particulate mobility, through the De Martonne Aridity Index (AI), was evaluated using the same meteorological data described for the GW Pathway Indicator characterization. The AI is estimated by the following formula:

$$AI = P \div (T + 10)$$

where: P is the mean annual precipitation (mm/yr) and T is the mean annual temperature (°C).

The assigned scores for the Particulate Mobility factor based on the AI are explained in Table 7. The AI in Campania region varies between 26 and 64, so the Air Particulate Pathway Indicator, represented by the Particulate Mobility factor, has only scores of 66 or 33.

The Air Particulate Receptor Indicator is evaluated by two factors: land use at site, and distance from the nearest residential area. The Corine Land Cover (2012) map was used for classifying the land use. From the same map the urban areas were extrapolated, and produced the Euclidean distance map through the ArcGIS spatial analysis module, which calculates the distance of each point (source) from the residential areas. The scores for both factors are indicated in Tables 8a and 8b.

The factor that mostly affects the Air Particulate exposure route map (Fig. 9) is the presence of residential areas. The highest scores (between 33 and 66) were found mainly in the Neapolitan province and on the Sarno River plain, and sparsely on the Lattari Mounts, Salerno urban area, Sele river plain and in south Cilento. Values between 23 and 33 were found in the same urban areas, in particular in the Campi Flegrei area and in the Campania plain. Some sparse value between 23 and 33 can be also found in the Avellino province, in Caserta province, from Alife to Trebulani Mounts (Mt. Maggiore) to Roccamonfina volcano (Sessa Aurunca), on the Volturno and Sele river plains and in south Cilento area. Scores between 13 and 23 are still in the main urbanized

Table 7
Scoring model for the Air Particulate Pathway Indicator based on the De Martonne Aridity Index values.

Aridity Index	Particulate mobility factor scores
<25	100
25–42	66
42–74	33
>74	0

Table 8a
Scoring model for the Air Particulate Receptor Indicator; land use at site factor.

Land use at site	Land use factor scores
Residential	60
Park/school/beaches	51
Agricultural/livestock	36
Industrial/commercial	26
Isolated areas	0

Table 8b
Scoring model for the Air Particulate Receptor Indicator; distance of the Source from the nearest residential area.

Distance from the nearest residential area (m)	Distance factor scores
<400	40
400–1070	27
1070–2200	20
2200–3350	14
>3350	7

areas, from the Neapolitan province to Caserta, Avellino and Salerno. The remaining inland territory, comprising mostly agricultural and isolated areas, has the lowest scores (0 − 13).

4.3.4. Direct Contact (DC) map

The DC exposure route map (Fig. 10) is obtained by means of the spatial analysis module in ArcGIS software, through the following expression:

$$DC_{er} = (S_{dc} * R_{dc}) \div 10^2$$

where: S_{dc} is the direct contact hazard map and R_{dc} is the DC Receptor Indicator.

For the DC exposure route the Pathway is not relevant, because it is assumed that the Source is coincident with the Receptor. What determines the vulnerability for the Receptor in this case is the type of land use and the distance from the Source. The Corine Land Cover map and the Euclidean distance map for the urban areas were used, and both reclassified according to the scores in Tables 9a and 9b.

In this case, the main factor conditioning the vulnerability of human health is the presence of residential areas near a potential contaminated site. Combining the highest scores of the hazard map and the DC factors, the DC exposure route with the main impact (having scores between 34 and 100) was obtained in the metropolitan area of Naples, and in the area between Avellino and Salerno provinces (Fig. 10).

4.4. Human health risk map

The overall human health risk map (Fig. 11) is computed by the root mean square of the four exposure routes, using the following algorithm (EEA, 2006):

$$R = \sqrt{\left(GW_{er}^2 + SW_{er}^2 + Air_{er}^2 + DC_{er}^2\right) \div 4}$$

where: GW_{er} is the Groundwater map, SW_{er} is the Surface water map, Air_{er} is the Air Particulate map, and DC_{er} is the Direct Contact map.

The map of the human health risk has been classified according to four classes of risk: very low, low, medium and high risk.

The PRA.MS methodology provides also an uncertainty analysis on the overall human health risk score, that reflects the quality of the data. The uncertainty analysis takes into account the influence in final site score of not available and not accurate input data, such as the use of geo-referenced data of low resolution. Assessments carried out with generic and low accuracy data may result in very high, and sometimes unacceptable, uncertainties (EEA, 2005b). The original calculations exposed in the PRA.MS for the uncertainty cannot be applied in this case, because geo-referenced data are considered. At a single industrial site it was possible to collect specific information for characterizing the parameters in a restricted area. In this case each pixel is considered to be a possible source. The uncertainty in such a case is not related to missing data, but is dependent only on the resolution of the geo-referenced data. In the Campania region risk assessment, the pixel resolution for all plotted maps is 20 m. The main uncertainties are related to: (a) the Boolean surfaces produced for the SW pathway indicator, due to the absence of flooding return periods information in the floodplains; (b) the assumption for the SW Receptor Indicator that the most common surface water use in all Campania region is the irrigation with food crops; (c) the use of the low-density meteorological information of the Regional Agrometeorological Network for the elaboration of the Air Particulate and (d) the GW Pathway Indicator maps. An implementation is required in a follow-up work for quantifying the uncertainties.

5. Results and discussion

The dominant exposure route is the Direct Contact, reaching scores up to 100 in the Neapolitan province, and in the area between Avellino and Salerno. This means that the main risk for the population in these areas comes from the direct contact with the soil that potentially contains pollutants. The risk is increased by the fact that the Neapolitan, Salerno and Avellino provinces have the higher number of residents, between 20,000 and 50,000 (Fig. 2). Moreover, intense agricultural and viticulture activities are diffused in the same areas. Secondary exposure routes, with scores reaching 75, are contamination of groundwater, surface water and diffusion in air. The overall risk map shows a similar situation, with the maximum risk, up to 63, restricted to the Neapolitan area, and mainly in the eastern area of the city. Moderate risk, with scores between 40 and 60, can be found in all the metropolitan area of Naples and province, from Campi Flegrei to Aversa, to Sarno river plain, and the area between Avellino and Salerno.

The aim of this research is to identify potential hazardous areas in the Campania region, and to prioritize them in order to use more efficiently the available resources for more detailed studies, and if needed their remediation. Potentially hazardous areas are defined in the PRA.MS as areas where the soil contamination does not affect only the local environment, but poses significant risks to human health and/or ecosystems (Quercia et al., 2006). Here there are conditions that allow the contamination to spread into the environment, and reach the population in different forms, such as inhaled dust or through the food chain. While the starting point of the PRA.MS is a preliminary identification of potentially hazardous areas, and the aim is their scoring, the starting point should be the chemical characterization of the regional territory in order to locate precisely the potentially hazardous areas. For these areas, there is the necessity to obtain more detailed data and information. Hence, the next step is high-density sampling and a follow-up risk assessment of selected areas.

The previous regional methodologies were based on selected industrial and mining sites (EEA, 2005b, Pizzol et al., 2011), resulting in a scoring of those sites, or on the effects of a single contaminant (Tristán et al., 2000). The new approach can help to obtain a multielement characterization of a whole regional territory. Each pixel on the maps shows a portion of territory, and, in the final risk map, indicates how much that territory is subjected to contamination. In order to obtain this, all the possible exposure routes have been taken into account. The quality and density of the initial data is fundamental for reaching an assessment as close as possible to reality.

On a wide and industrialized territory, such as the Campania region, is very difficult to understand where to install monitoring systems. Some areas with a score of 75 on the hazard map become, in the final risk map, areas with low risk. This means that the application of the methodology and the elaboration of the overall risk map do not change the general distribution of the hazard, but narrow the limit of the areas of interest, giving a first contribution for the identification of priority areas.

The risk assessment has identified two areas that can be characterized as "potentially hazardous". The first one is the Neapolitan area, which has been the subject of many previous published works, with detailed sampling and characterizations (De Vivo et al., 2006b; Cicchella, 2000; Cicchella et al., 2003, 2005, 2008; De Vivo and Lima, 2008). The second is the area between Avellino and Salerno, which is part of the agricultural zone with a low soil sampling density. The high population density, the presence of viticulture activities, with intense use of

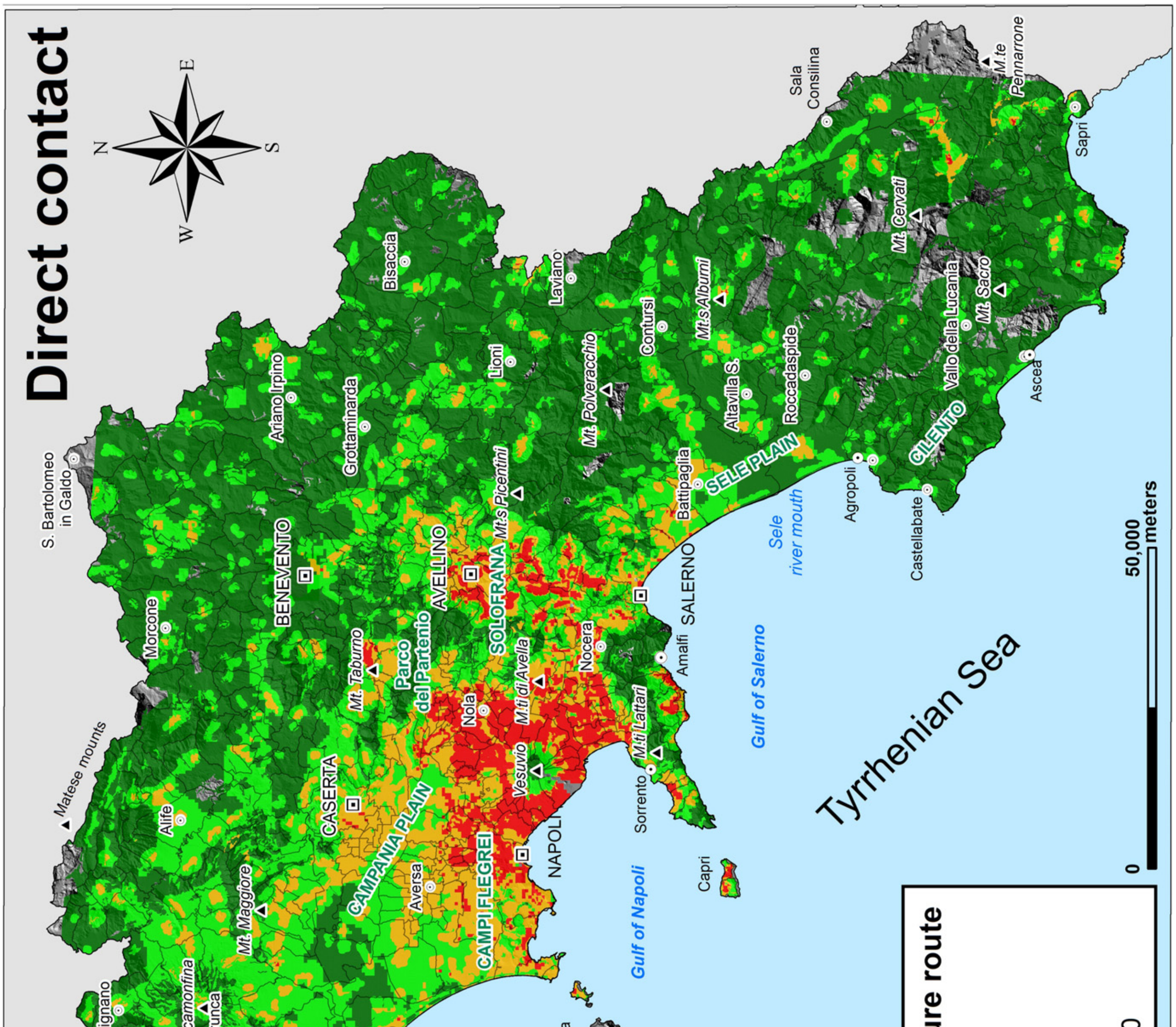

Fig. 10. Direct Contact (DC) exposure route map for Campania region.

Table 9a
Scoring model for the DC Receptor Indicator; land use at site factor.

Land use at site	Land use factor scores
Residential	47
Park/school/beaches	39
Agricultural/livestock	28
Industrial/commercial	19
Isolated areas	0

Table 9b
Scoring model for the DC Receptor Indicator; distance of the source from the nearest residential area.

Distance from the nearest residential area (m)	Distance factor scores
<400	53
400–1070	38
1070–2200	29
2200–3350	19
>3350	8

fertilizers, and the existence of industries (tanneries that are the main cause of one of the most contaminated sites in Italy, the Solofrana and Sarno River), make this area "potentially hazardous", as shown by the results of the regional risk assessment, and the map Fig. 12 (modified from Quercia et al., 2006). Industrial and agricultural wastes can seriously affect the groundwater, especially in an area with a high number of drinking water supply wells; due to the high slopes, the surface water bodies, which flow to one of the most important rivers of the Region (Sarno river), can carry diluted contaminants and, of course, a high number of residential population is clearly exposed to the contaminated media.

The identified potentially contaminated area from this regional risk assessment is still too wide to be directly used by the administrations for planning any environmental reclamation and rehabilitation. However, the methodology is useful to identify well-suited areas for a follow-up surveys by increasing the sampling density. Then the same risk assessment methodology described here, can be applied in each area. The denser sampling in the smaller area will provide more precise information about the human health risk, and can be helpful in the planning of follow-up steps. The developed risk assessment methodology should be applied in each area, as the objective is to give to the administrations a valid tool in the decision-making process with respect to land use. The risk map showing the distribution of potentially hazardous areas, should simplify the decision process about the end land use of each area. Another possible advantage of having a regional mapping of environmental risk is the application in epidemiological studies, already carried out by Albanese et al. (2013). They found that there is a good spatial correlation between the incidence of some cancer types and the distribution patterns of contaminants in stream sediment. The use of the regional risk map would be more appropriate than the single element distribution maps, as it provides more complete views of the environmental and sanitary risk.

Fig. 11. Overall risk map for human health risk assessment in Campania region.

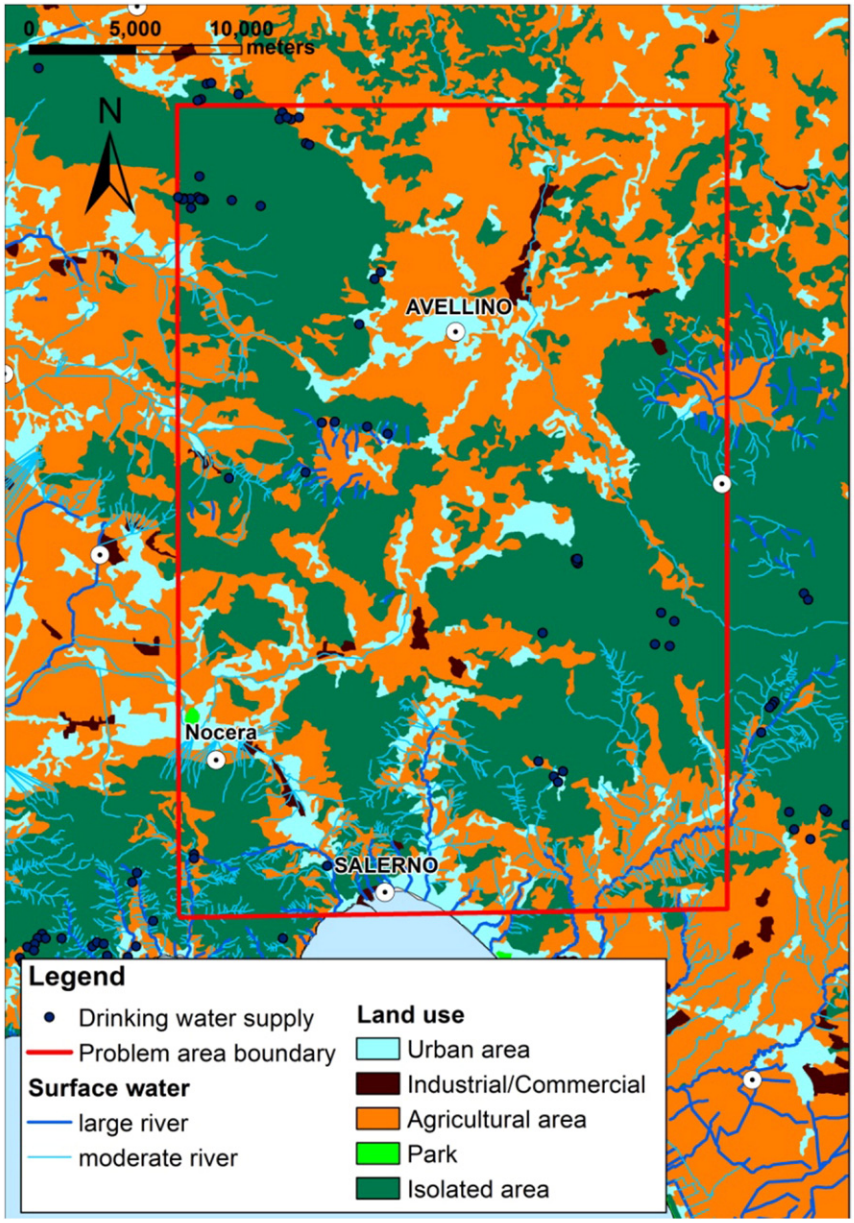

Fig. 12. Potential hazardous areas, located between Avellino and Salerno provinces.

6. Conclusions

The methodology proposed in this paper provides a new approach for assessing the risk for human health in intensive and extensive contaminated regions. A European-wide accepted methodology for the preliminary assessment of human health risks at single contaminated sites (PRA.MS) is the base for this study. In order to adapt this methodology to the regional level, GIS software tools were used in order to evaluate effectively the spatial distribution of different parameters. The methodology consists in a multielement characterization of the soil in a regional territory, where all possible exposure routes for contamination are evaluated.

The regional approach is useful for the administrations' decision-making process to select the potentially hazardous areas, and to use cost-effectively the available resources. A fundamental step in guaranteeing the protection of soil, and consequently the health of the population, is focusing the monitoring and action systems on selected potentially hazardous areas, so that the monitoring efficiency is increased and the costs are minimized. Every kind of decision-making process regarding the environmental and population health would be simplified through the elaboration of a regional risk distribution map.

The main difficulty of carrying out a regional risk assessment is the availability of data at this level. The proposed method can be improved by the collection of more detailed and accurate data and information, and the quantification of uncertainty. After selecting the potentially hazardous areas it is possible to apply the same methodology in a more restricted area, where more parameters are available, so that the accuracy of the risk assessment will be better.

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