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Agricultural areas in potentially contaminated sites: characterization, risk, management

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Abstract

Introduction. In Italy, the current legislation for contaminants in soils provides two land uses: residential/public or private gardens and commercial/industrial; there are not specific reference values for agricultural soils, even if a special decree has been developed and is currently going through the legislative approval process. The topic of agricultural areas is relevant, also in consideration of their presence near potentially contaminated sites.

Aim and results. In this paper, contamination sources and transport modes of contaminants from sources to the target in agricultural areas are examined and a suitable "conceptual model" to define appropriate characterization methods and risk assessment procedures is proposed. These procedures have already been used by the National Institute of Health in various Italian areas characterized by different agricultural settings.

Conclusion. Finally, specific remediation techniques are suggested to preserve soil resources and, if possible, its particular land use.

Key words
- agriculture
- soil
- risk assessment

INTRODUCTION

Italy is considered a country with a strong agricultural vocation, nevertheless the loss of agricultural land is a constantly growing phenomenon, also due to the proximity of these areas to contaminated sites. The potential contamination of the land is caused by both point and diffuse pollution sources. The contamination is caused by point sources if it involves limited areas; point sources can be industries, harbours and landfills. Nevertheless, the release in the environment of large amounts of chemicals by industrial, civil (vehicular traffic), and agricultural (incorrect use of pesticides, without attention to the effectiveness doses and the latency periods) activities, can generate a diffuse pollution, involving wider areas. In this case, the contamination is also due to the potential rainwater runoff and atmospheric transport of contaminants, with ground deposition also far away from the emission source. Finally, potential water contamination, both deep and surface, can transfer contaminants to the soil through irrigation.

In some European countries, legislative reference values for agricultural soils are provided; these soil screening values are generic quality standards used to regulate land contamination [1]. The Italian environmental legislation does not provide chemical concentration values to assure the quality of agricultural soils. The current legislation, Legislative Decree Framework n. 152/06 [2] considers only two soil uses: residential/public or private gardens and industrial/commercial. As regards the agricultural areas, the article n. 241 of the decree refers to a future regulation on remediation, environmental restoration, emergency securing, both operative and permanent, to be taken by the Ministry of the Environment in collaboration with ministries of Productive Activities, and Health, Agricultural and Forestry Policies. This decree has not yet been adopted.

Recently new rules, such as the Law n. 6 of February 6, 2014, were implemented and the competent ministries worked at drafting the agricultural areas decree, that is still not legally completed. This follows the contamination events of the "Terra dei Fuochi" area in Campania region, that have generated a strong media alarm. The decree has a focus on the development of methods for characterization, risk assessment and management, with the aim of minimization of the related potential risks and has the objective to retrain the same areas, where it is necessary and/or possible.

In order to carry out a health risk assessment in a contaminated area, it is necessary to develop a Conceptual Site Model (CSM). It is a representation of the environmental system and of the physical, chemical and biological processes, that determine the transport of contaminants from sources to receptors [3]. A CSM generally includes information about contamination

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sources, transport pathways, exposure pathways and receptors. To reach this aim, it is necessary to carry out an accurate site characterization, studying the history of the area and verifying the presence of industrial plants or other production systems currently ongoing or active in the past. It is important, also to check the presence of incineration and waste disposal plants. Different analyses should be carried out such as surveys piezometers placement, chemical analyses and other studies in order both to define the geological and hydrogeological system and to check the contamination in different environmental media. After that a correct CSM has been developed, a suitable characterization plan of the area is planned.

CHARACTERIZATION IN AGRICULTURAL AREAS

In a contaminated site, including also agricultural areas, the CSM can be represented by two distinct linked patterns (*Figure 1*): an environmental conceptual model, where the target is an environmental matrix and a health conceptual model, where the target is the general population [4]. In the environmental conceptual model, the contamination sources are due to anthropogenic activity, such as industrial or vehicular emissions, dust deriving from excavation or materials transport. It is important also to control dump or waste disposal areas. In agricultural areas, the massive or improper use of plant protection products could be a problem too. The target is the environment, that is agricultural or grazing areas. The contaminant transport from sources to receptors can occur through atmospheric dispersion and/or powders relapse; for this reason it is significant to study the intensity and direction of the prevalent winds. A further contribution to the contamination could be caused by rainwater permeation or runoff and by use of contaminated irrigation water.

The conceptual health model is derived from the environmental one; the target of the environmental model, that is agricultural areas, becomes the contamination source of the health model. The receptor is the entire population and the exposure is indirect. The transport occurs mainly by dietary intake, through consumption of potentially contaminated foodstuffs produced in the same area.

A characterization plan should be defined in order to study agricultural areas,; it should be able to describe the soil properties, in particular those related to its vulnerability and the potentiality to lose one or more of its ecological functions [5]. The presence of the contaminants should be investigated in all environmental media (water, air and soil). For a particular group of substances such as heavy metals, the bioavailable form could be investigated. The plan should be able to provide recommendations about a possible translocation of toxic elements in agricultural products, because the vulnerability of agricultural soils is related to vulnerability and quality of agricultural food production.

In the definition of the environmental conceptual model, some contaminants are identified as "index pollutants". They are representative of the area contamination because of their frequency detection, the high levels of concentrations, and their chemical, physical and toxicological properties [6].

In agricultural areas the main matrices to be characterized are soil, fruits and vegetables.

Sampling

As regards sampling of agricultural soils, the site should be distinguished in homogeneous areas. If homogeneity degree is unknown, official methods of physical soil analysis (geophysical field surveys by electromagnetic induction or electrical resistivity) are used [7]. These surveys need to highlight the homogeneity/heterogeneity degree due both to natural and anthropogenic causes, such as the various properties of the soils or the presence of allochthonous materials (landfill waste). As a second step, a map of homogeneous areas is carried out. To characterize these areas, a different sampling procedure [8], compared with the one used

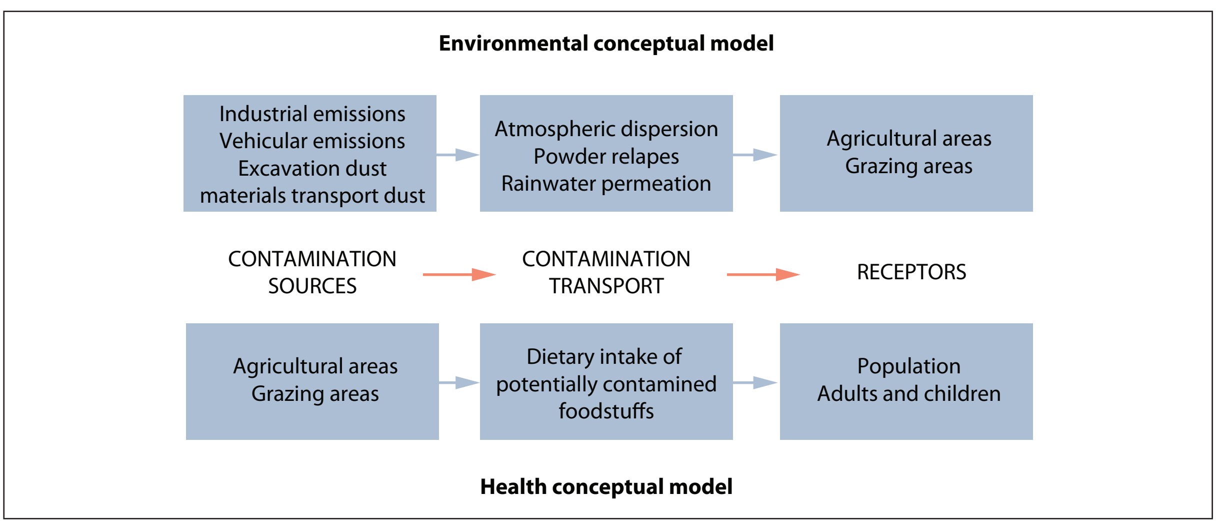

Figure 1
Conceptual model in agricultural areas.

for other destination uses, is used. Generally, in a contaminated site (residential/public or private gardens and industrial/commercial), the soil sampling is carried out by coring. In agricultural areas, a frequent soil mixing is present, therefore a "bulk sample, composed by mixing several (incremental samples), is considered more representative". The "incremental samples" are taken in a land plot, considering a different sampling depth according to various crops. "Incremental samples" should be kept to make an in depth investigation, if it becomes necessary.

Regarding the water quality, the Italian legislation provides specific quality parameters and standards for irrigation only for wastewaters [9]; water analyses (in surface and groundwater bodies) should be carried out to verify potential correlations with soil pollution.

This kind of sampling procedure, as the following risk assessment procedures, has already been carried out by the National Institute of Health in several different Italian areas, like the National Contaminated Sites of Sulcis in Sardina region (small family allotments) and of Brescia Caffaro in Lombardia region (a large, mainly agricultural area) [10, 11], and the Giugliano wide Area in Campania.

It is important to remark that the characterization may include monitoring foodstuffs plans, but this is considered a next phase in the risk assessment procedure.

RISK ASSESSMENT IN AGRICULTURAL AREAS

The Legislative Decree n. 152/06, uses the "Risk Analysis" as a decision support tool for the risk assessment and management in residential/public or private gardens or industrial/commercial soils, performed by mathematical-probabilistic standard procedures. It determines Threshold Risk Concentrations (TRC), that are remediation goals for the site (backward method) and returns the estimate of the health risk for human receptors (forward method). If a standard Risk Analysis was carried out in agricultural soils through the backward method, as required by the current legislative framework, humans should be the target and the human receptor would be the farm worker, that is subject to a professional exposure. For this reason, a Risk Analysis by backward method is not appropriate, because it would lead to a TRC exclusively aimed at the farm worker protection; however it is possible to develop a forward Risk Analysis in order to estimate the professional risk and to provide for the use of Individual Protection Devices (IPD).

In agricultural areas, it is appropriate to use criteria and assessment methods different than those described above for other land uses (*e.g.* residential), because the potential contamination may pose a risk to public health, due to the possible soil-plant transition and the following entry into the food chain.

The risk assessment procedure can be divided into different steps; the priority should be the enhancement of the characterization of the area. It is known that the risk assessment based only on the control of total contaminants content in soil does not provide any information about their mobility and bioavailability. To assess properly the risk related to the soil contamination and to predict its attenuation following the application of proper remediation techniques, the contaminants mobility and bioavailability in different climatic conditions should be evaluated, through the use of suitable analytical methods. Generally, analytical determinations required for bioavailability assessment are texture, pH, redox potential, organic carbon, cation exchange capacity, carbonate content. Analytical tests to evaluate bioavailability give information about possible contaminants translocation in fruit and vegetables; they are relevant if there is no possibility to carry out crops monitoring (*e.g.* agricultural areas where the use is forbidden). For a more thorough characterization, suitable monitoring fruit and vegetables plans are carried out. Defining the accumulation of pollutants in the edible parts of plants grown in different soils and various agronomic management allows to identify the potential hazard to humans and to animals. If there are pasture lands or forage crops, it may be appropriate to monitor also foods of animal origin (milk, meat, eggs), because the contamination can reach them through contaminated fodder consumed by the animals.

According to site-specific characteristics crops in the area, one or more food matrices can be sampled. If a particular "index pollutant" (*e.g.* plant protection product) has been identified in the conceptual environmental model and if its specific use of a foodstuff is known, monitoring data can be limited to the same crop. On the contrary, in a site next to industrial areas, where more than one "index pollutants" have been identified or predicted, it is correct to carry out a monitoring as various as possible. It is necessary to consider all the foodstuffs grown in the area, in order to assess the potential contaminants intake from all foods, also due to the seasonal variability. Where it is possible, crops sampling should be performed next to the soil sampling.

Different approaches in risk assessment

In the assessment procedure, it is necessary to collect a statistically significant number of samples with a good quality for analytical purposes. It is required a prior critical data evaluation, to assess their reliability and comparability; they also need to be harmonized in order to express the final result.

For many chemicals, there are not legal limits on foodstuffs, therefore risk thresholds related to their intake should be defined. The risk assessment related to food consumption may be carried out through different approaches. Data on food consumption are available in periodic studies performed by the National Institute of Research on Food and Nutrition (ex INRAN), now part of the Council for Research in Agriculture- Research Centre for food and Nutrition (CRA-NUT) [12]. These data are also available depending on the geographic distribution, age and gender of consumers. They also are included in the European food consumption database of the European Food Safety Authority (EFSA). A comparison between the contamination intake through diet and tolerable daily doses (*e.g.* Tolerable Daily Intake TDI, Acceptable Daily Intake ADI) suggested by scientific accredited organizations (*e.g.* WHO, EFSA) is carried out. These doses can also be considered temporary (*e.g.*Provisional Tolerable Weekly Intake, PTWI), considering the available evidence. If toxicological parameters as TDI, ADI, TWI are not available, the risk assessment can be carried out through the US Environmental Protection Agency (EPA) approach, like the Risk Analysis performed by mathematical-probabilistic standard procedures. This method uses the Reference Dose (RfD) for the assessment of toxic effects and the Slope Factor (SF) for carcinogenic ones, furthermore it considers parameters as Exposure Frequency (EF) and Exposure Duration (ED), to which more or less precautionary values can be attributed. Even the food daily dose (Intake Rate IR), used in the calculation formulae to estimate contaminants daily intake, can have different values. The daily intake rate can be considered fully or partially made up of agricultural products from the area; considering different types of local production (*e.g.* family gardens or large production), different evaluations can be performed by the risk-assessors.

MANAGEMENT OF AGRICULTURAL AREAS TO BE REMEDIATED

If agricultural soils are not more suitable for agricultural land use, safety and/or remediation actions are needed. The goal is to preserve the soil resources, therefore it is essential to reduce remediation activities such as removal, transportation, excavation and washing actions; these techniques could be used when other *in situ* and smaller impact strategies result insufficient. The ecosystem balances, that led to the soil formation, need to be maintained in order to return to the traditional agricultural use of the soil, as quickly as possible.

For agricultural areas clean-up, phytoremediation and bioremediation techniques are preferred rather than chemical-physical treatments. These methods are cheaper and have a lower environmental impact and are more acceptable to public opinion.

Phytoremediation techniques include various processes; some of them act mainly on inorganic contaminants while others act mainly on organic compounds [13]. For inorganic contaminants the phytoextraction is applied. It is based on the uptake of contaminants in the soil by plant roots and translocation within the plant. Even rhizofiltration, the absorption of contaminants by roots, is used, but roots should be in solution surrounding the root zone. A third technique is the phytostabilization based on the immobilization of contaminants in soil through reducing their bioavailability. For organic compounds, phytodegradation and phytovolatilization are preferred; phytodegradation is the breakdown of contaminants taken up by plants through metabolic processes, while phytovolatilization is the uptake and transpiration of contaminants by plants, with their release (even in modified form) to the atmoshere [14].

As regards bioremediation, various techniques are used; for example the bioaugmentation involves the addition of supplemental microbes (bacterias and fungi) to the soil where organisms are not able to degrade specific contaminants, while the biostimulation involves the addition of key biological building blocks, such as nitrogen and phosphorus and other trace nutrients necessary for cell growth [15], thereby the native microflora metabolism is increased.

If the soil contamination is too high, more effective approaches can be used, as chemical (oxidation, reduction, soil-flushing, fixation) or physical treatments (solidification, thermal actions).

Other factors should also be taken into account to assess the health risk and to select the best appropriate technique; for example, in the case of metal pollution, the bioavailable fraction has a key role because only this fraction is subject to the mechanism of absorption to the crops and mobilization in the deep parts of soil and subsoil. Moreover, various parts of the plants (leaves, stems, fruits, seeds) [16] can absorb contaminants in different amounts; for this reason, in the risk assessment stage it is necessary to consider the edible parts that allow the contamination transfer in the food chain, while in the remediation stage it is right to use plants that can absorb contaminants through more suitable parts.

CONCLUSIONS

Considering their particular use, agricultural areas (especially within or next to contaminated sites) need specific characterization and risk assessment procedures to be used for management and remediation activities. As regards characterization, firstly it is preferred to sample soil by "bulk sample" rather than by coring; secondly, it is important to carry out a monitoring of fruits and vegetables, to assess the health risk related to the potential intake of contaminants through the diet. In calculating risk, various exposure factors are considered and they can take different values (more or less conservative); site-specific considerations allow the best choice. Moreover, since it is not possible to take measures to completely eliminate risk factors, a careful risk-benefit analysis, aimed to the health protection and to the food safety production, must be always carried out.

Regarding to safety and remediation actions, it is of prime importance to preserve the soil resource, so *in situ* phytoremediation and bioremediation techniques are preferred.

All these actions should contribute to a general policy of health and environmental protection; in this respect, it is very important the risk communication to the population. This allows to achieve a collaboration with the end users, without arising alarmism and educating the population to correct behavior.

Conflict of interest statement

The authors declare that they have no conflicts of interest.

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