Ann Ist Super Sanità 2010 | Vol. 46, No. 3: 303-308
DOI: 10.4415/ANN_10_03_14

Agricultural soils potentially contaminated: risk assessment procedure case studies

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Summary. At the moment, the health-environmental risk analysis is used to decision-making targets in the contaminated sites management; this procedure allows to assess the quantitative health risk related to the pollutants presence in environmental compartments, as soil and waters. As regards potentially contaminated agricultural soils, the ingestion of food from vegetable and/or animal source, produced inside the contaminated area, is the most suitable way to assess the health risk. As an official procedure to this assessment is not available, the National Institute for Health (Istituto Superiore di Sanità, ISS) has worked out an operating procedure, organized into several phases, depending on the available specific-site know-how. In this document, agricultural soils potentially contaminated in two sites have been studied; the sites are the following: Brescia Caffaro and Torviscosa.

Key words: soil, agriculture, risk assessment.

Riassunto *(Suoli agricoli potenzialmente contaminati: casi studio di procedure di valutazione del rischio)*. Attualmente, l'analisi di rischio sanitario-ambientale è lo strumento utilizzato, per fini decisionali, nella gestione dei siti contaminati; essa permette di valutare quantitativamente i rischi per la salute umana connessi alla presenza di inquinanti nei diversi comparti ambientali, quali suolo e acque. Per i suoli agricoli potenzialmente contaminati, si ritiene che il mezzo più idoneo per la valutazione del rischio sanitario sia la stima dei consumi di alimenti, di origine vegetale e/o animale, prodotti all'interno dell'area contaminata. Non esistendo, al momento, una procedura ufficiale di valutazione, l'Istituto Superiore di Sanità (ISS) ha messo a punto un protocollo operativo, strutturato in diverse fasi, secondo le informazioni sito-specifiche disponibili. In questo articolo, sono stati studiati suoli agricoli potenzialmente contaminati situati in due diverse aree: Brescia Caffaro e Torviscosa.

Parole chiave: suolo, agricoltura, valutazione del rischio.

INTRODUCTION

In a contaminated area the main purpose of investigating different matrices is the assessment of the nature and extent of contamination, in order to determine if there are unacceptable risks for human health and to evaluate potential remedies. At the moment, the health-environmental risk analysis is used to decision-making targets in the contaminated sites management. A risk analysis must be preceded by a site characterization process, that allows to determine and quantify the contaminants present in different environmental compartments, such as soil and waters. Moreover, it is important to identify the sources of historical and/or present contamination and to understand the processes affecting the environmental fate of contaminants. At the end, more complete human and ecological exposure pathways should be known.

Concerning the risk analysis of industrial/commercial or residential areas, there are standardized procedures which make use of commercial softwares; they consider routes of exposure such as inhalation and dermal contact.

For agricultural soils, the ingestion of food from vegetable and/or animal source produced inside the contaminated area is considered the most suitable way to assess the health risk. As an official procedure to this assessment is not available, the Italian National Institute for Health (Istituto Superiore di Sanità, ISS) worked out an operating procedure, organized into several phases, depending on the available specific-site know-how.

As shown in the *Figure 1*, at first contamination sources of the agricultural areas should be identified; they can be the agronomic practices or other sources as, for example, working or inactive industry. Then a comparison with regulation values for pollutants on soil should be carried out.

In some European Countries legislative values for agricultural soils are provided; these soil screening values are generic quality standards that are used to regulate land contamination [1].

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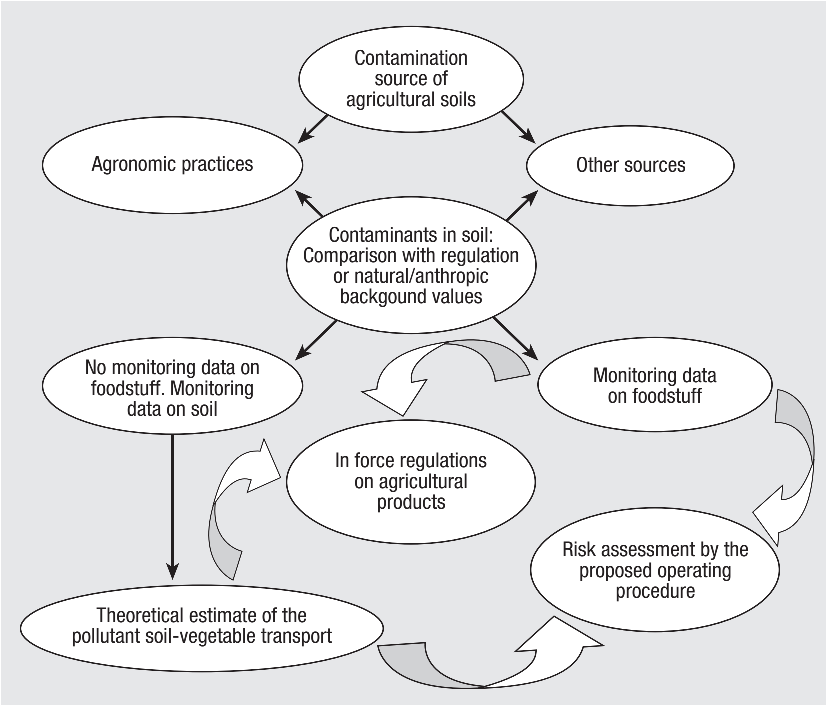

Fig. 1 | *Health risk assessment approach in agricoltural soils.*

In Italy legislative values for agricultural soils are not provided, so a comparison with natural or anthropogenic background should be carried out; however, if the presence of the contaminants in soil is suspect, it is appropriate to run a monitoring of the area.

If the background values on soil are suspected to exceed guideline levels or to be, however, too high, but it is not possible to conduct a monitoring programme and there are not previous analytical data, theoretical estimates of the pollutant soil-vegetable transport could be made from concentrations in foodstuffs. Then, comparison with legislative levels for foodstuffs – *e.g.* comparison with maximum residue level (MRL) for pesticides – or with monitoring data on foodstuff should be performed.

In both cases it is possible to carry out a risk assessment according to the proposed operating procedure described later.

Likewise to the risk analysis carried out by the softwares, the starting point for the health risk assessment in agricultural areas is the development of a conceptual site model (CSM). The CSM 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. A CSM generally includes information about contamination sources, transport pathways, exposure pathways and receptors.

In the case of vegetables, there are different contamination pathways: radical absorption, and volatilization from soil and atmospheric deposition, that is the most important one [2].

In the specific model by food ingestion [3], the most important way of the exposure in the agricultural areas, the source corresponds with the site investigation, *i.e.* the studies allowing to identify the index-pollutants. Index-pollutants are the most representative pollutants in a site for their high concentration and/or their toxicological characteristics.

The transport corresponds to the monitoring of vegetable and/or animal origin food and to the food intake rate; adults and children are the target, for which a risk assessment by toxic and/or carcinogenic substances is carried out.

OPERATING PROCEDURE

The proposed operating procedure for risk assessment is organized into several phases:

1) estimation of the concentration level of pollutant index in food products through:
   a) collection and selection of monitoring data on foodstuffs or soil and following statistical processing;
   b) theoretical estimate of the pollutant soil-vegetable transport by biotransfer factor;
2) estimation of food intake *pro capite* (intake rate, IR) and contaminant quantity taken daily by diet $[\Sigma_i(C \times IR)_i]$, when there are not legislative values for the pollutant index;
3) use of average daily intake:
   a) average daily dose (ADD) for toxic effects; and/or
   b) lifetime average daily dose (LADD) for carcinogenic effects;
4) risk calculation: average daily intake supplemented with the specific toxicological parameters of substances:

a) for toxic effects: HI = ADD/RfD (reference dose, acceptable intake);
and/or
b) for carcinogenic effects: R = LADD×SF (slope factor, value, in inverse concentration or dose units, derived from the slope of a dose-response curve, limited to carcinogenic effects with the curve assumed to be linear at low concentrations or doses).

Since a monitoring on foodstuff is the best way to explain the effective contamination level of the area, the use of real data (phase 1.a) is recommended than the theoretical estimate of the pollutant soil-vegetable transport by biotransfer factor (phase 1.b). The analytical data should be reliable and comparable, and the number of available analyses should be suitable to allow a statistical processing.

If there are not analytical data and it is not possible to plan a monitoring programme, a theoretical estimate of the concentration level of the contaminants on vegetables can be carried out.

In the mentioned phase 2, the food intake *pro capite* (intake rate, IR) is estimated and it is possible to use studies carried out by Italian National Research Institute for Food and Nutrition (INRAN) [4-6]. These studies, conducted over the years, generally presents the main results of the Italian national food consumption surveys. The results are explained by food category; in each category different products are included, *e.g.* cereals, cereal products and substitutes included bread, pasta and pasta substitutes; pizza, rice, wheat, etc.; fresh fruit included apricots, apples, pears, etc. The food intakes are shown not only in the total population, but also by gender, age class and geographical area. After the estimation of IR, for the analytical data of each food category, a statistical processing is carried out to choose a representative Concentration value (C) that is a concentration value that is significant of the contamination level for each food category.

This value can be obtained by arithmetic mean, by half detection limit (if all the analytical results are smaller than the detection limit) or by the use of ProUCL software (www.epa.gov/nerlesd1/tsc/software.htm), a statistical one that computes upper confidence limit (UCL) of the unknown population mean. Than, the contaminant quantity taken daily by diet $[\sum_i(C\times IR)_i]$ can be assessed.

In the phase 3, the average daily dose (ADD, mg/kg day) for toxic effects and/or the lifetime average daily dose (LADD, mg/kg day) for carcinogenic effects is calculated using the following equation [7]:

$$\text{ADD or LADD} = \frac{\sum_i (C\times IR)i \times EF \times ED}{BW \times AT}$$

where:
- Ci is the representative concentration value of the index-pollutant in each food category (mg/g);
- IRi is the intake rate for each food category (g/day);
- EF is the exposure frequency (days/year);
- ED is the exposure duration (years);
- BW is the average body weight of the receptor (kg);
- AT is the averaging time, the period over which exposure is averaged (days).

The difference between ADD and LADD is the AT value as described in the Brescia-Caffaro site description.

In the phase 4 the risk calculation is carried out, supplementing ADD and/or LADD with the specific toxicological parameters of substances, Reference Dose (RfD) for toxic effects and slope factor (SF) for carcinogenic effects.

If several index-pollutants are determined, the assessment procedure is applied separately for each pollutant.

AGRICULTURAL CONTAMINATED SOILS

The Brescia-Caffaro site

In the Brescia-Caffaro site the index-pollutants are the polychlorinated biphenyls (PCB). PCBs constitute a group of 209 congeners, twelve of them show toxicological properties similar to the dioxins (PCB dioxin-like). There were previous analytical data provided by the Brescia Local Health Company, so the phases 1.a and 2 have been applied.

Phase 1.a

In the analytical data set there were several congeners; all the investigated congeners were considered as total PCB (PCB_{tot}).

The different available matrices have been collected in food categories according to the INRAN studies: dried fruit (dried figs, almonds, walnuts), fresh fruit (apricots, sour cherries, apples, figs, pears), garlic, onions, thistles/carrots/fennels/turnips/celery (beets, carrots, leeks, radishes, celery), spices (bay leaves, origanum, parsley, rosemary), cauliflower (cabbage, savoy), potatoes, cucumbers/courgettes, aubergines/peppers, tomatoes, salads (salad, lettuce, chicory), vegetables (green beans, spinachs), legumes (beans, peas). Then a statistical processing has been carried out to choose the C.

In *Table 1* food categories, available analytical data, statistical procedures for choosing C, and the resulting C values are shown.

Phase 2

IR has been estimated according to the INRAN studies and contaminant quantity taken daily for each food category has been assessed (CxIR). The total contaminant quantity taken daily by diet $[\sum_i(C\times IR)_i]$ is 2.24×10^{-3} mg/day *pro capite (Table 1)*.

Phase 3.a. and 3.b

In the phase 3 the average daily intake assessment has been applied separately for adults and children.

In this phase ADD and LADD have been both calculated because the presence of a PCB Dioxin-like cannot be excluded (see note 5 of ISS-ISPESL database).

The used parameter value are 350 days/year for EF, because a fifteen day holiday period has been considered; as regards ED, the real total time period

Table 1 | *Information about the analytical data on the food categories*

Food category	Available analytical data	Statistical processing	Representative concentration value PCBtot (µg/kg)	Representative concentration value PCBtot C (mg/g)	Food intake rate IR (g/day)	Contaminant quantity daily taken by diet (CxIR) (mg/day)
Dried fruit	3	1/2 D.L.(1)	2.00	2.00E-06	2.9	5.80E-06
Fresh fruit	5	1/2 D.L.(1)	2.00	2.00E-06	156.8	3.14E-04
Garlic	3	1/2 D.L.(1)	2.00	2.00E-06	0.7	1.40E-06
Onions	11	1/2 D.L.(1)	2.00	2.00E-06	8.8	1.76E-05
Thistles, carrots, fennels, turnips, celery	19	ProUCL	8.72	8.72E-06	16.1	1.40E-04
Spices	117	ProUCL	8.49	8.49E-06	0.4	3.40E-06
Cauliflower	5	Arithmetic mean	3.80	3.80E-06	10.3	3.91E-05
Potatoes	2	1/2 D.L.(1)	2.00	2.00E-06	54.5	1.09E-04
Cucumbers, courgettes	25	ProUCL	7.17	7.17E-06	12.3	8.82E-05
Aubergines, peppers	3	1/2 D.L.(1)	2.00	2.00E-06	15.2	3.04E-05
Tomatoes	17	ProUCL	1.17	1.17E-06	30.4	3.56E+05
Salads	62	ProUCL	37.22	3,72E-05	4.7	1.75E-04
Vegetables	45	ProUCL	93.09	9.31E-05	13.5	1.26E-03
Legumes	3	1/2 D.L.(1)	2.00	2.00E-06	13.9	2.78E-05

(1) *Detection limit, 4 µg/kg.*

over which contacts occur between receptors and contaminated foodstuff by the diet, a seventy year precautionary value can be used, but, in Brescia-Caffaro site, the value for ED is 40 years for adults, as certified by the Brescia Local Health Company document [8] and 6 years for children; BW is 70 kg for adults and 15 kg for children.

The AT values are different for ADD and LADD: in ADD (for toxic effects), the averaging time is the exposure duration expressed in days, therefore AT value is 14 600 days for adults (40 years × 365 days/year), whereas AT value is 2,190 days for children; in LADD (for carcinogens), the averaging time is based on a lifetime exposure of 70 years, therefore the AT value is 25 550 days for adults and children. All the exposure parameter values are obtained from US EPA-1997 and US EPA-2002 [7, 9].

ADD (mg/kg day) is 3.07×10^{-5} for adults and 1.43 × 10-4 for children; LADD (mg/kg day) is 1.76×10^{-5} for adults and 1.23×10^{-5} for children.

Phase 4.a and 4.b

In the phase 4, the risk calculation has been carried out. For toxic risk, the toxicological parameter is the RfD and the specific used value for PCB_{tot} is 2×10^{-5} mg/kg day, as referred in the database of ISS-ISPESL (www.apat.gov.it/site/it-IT/temi/siti_contaminati/Analisi_di_rischio); than the hazard index (HI = ADD/RfD) was calculated. HI is 1.54 for adults and 7.17 for children. For toxic effects the risk is acceptable if HI < 1.

For carcinogenic effects the toxicological parameter is the SF and the specific used value for PCB_{tot} is 2 (mg/kg day^{-1}) [11]; than the risk (R = LADDxSF) was calculated. The slope factor and the exposure is taken to reflect the probability of producing the related effect. R is 3.5×10^{-5} for adults and 2.5×10^{-5} for children. For carcinogenic effects the risk is acceptable if $R < 1 \times 10^{-6}$. Even if the results obtained by the proposed operating procedure show risk for adult and children, it is important to notice that the estimate is precautionary. In fact, it has been considered that all the food consumption comes from the contaminated agricultural area, and the intake rate is the same for adults and for children. Moreover, since some of the investigated congeners were dioxin-like, the carcinogenic effects have been considered and the risk has been calculated as indicated by the procedure; nevertheless most congeners were non dioxin-like and the EFSA Panel (2005) concluded that non dioxin-like PCBs are neither genotoxic or carcinogenic.

The Torviscosa site

In the Torviscosa site (in province of Udine, North Italy), the index-pollutant is Dieldrin. Dieldrin is a metabolite of Aldrin as well as a pesticide marketed in the past. They have been widely used in the past, but are now banned in most Countries world wide. Nevertheless, there are still residues of Dieldrin in the environment, even if the levels have been declining during the last 30 years. The fodder is the main growing in the Torviscosa area.

Phase 1.a

Seventy analytical results of Dieldrin concentration in soil, coming from the site characterization process, were available. Two different statistical processing of the analytical data have been carried out, in order to choose the representative concentration value in soil. At first the UCL was calculated and the corresponding Dieldrin representative concentration value in soil ($C_{soil\text{-}UCL}$) was 0.034 mg/kgsoil.

Since the maximum level in soil is the most precautionary value, in the analytical data set of Dieldrin concentration in soil, this value has been also considered. The Dieldrin concentration value in soil ($C_{soil\text{-}max}$) was 0.047 mg/kgsoil.

Phase 1.b

A theoretical estimate of the pollutant soil-vegetable transport has been carried out by the use of biotransfer factor, using the following equation:

$$C_v = C_{soil} \times B_v \times (1-0.12)$$

where:

- C_v is the Dieldrin estimated concentration in fodder (mg Dieldrin/kg vegetable);
- C_{soil} is the Dieldrin concentration in soil (mg Dieldrin/kg soil);
- B_v is the biotransfer factor from soil (mg Dieldrin/kg vegetable per mg Dieldrin/kg soil);
- (1−0.12) is an adjustment factor for humidity, considering humidity about 12%, as generally considered for feedstuff.

The environmental fate of organic chemicals is largely determined by their partitioning tendencies among aqueous, atmospheric and organic phases. The key parameter in assessing and describing the partitioning behavior of organic chemicals in the latter phase is the n-octanol/water partition coefficient (K_{ow}). So the B_v was calculated using the following equation [10]:

$$B_v = 10^{\,1.588-0.578 \log Kow}$$

where log $K_{ow\ djeldrin}$ is 5.37 [5]; so the calculated B_v Dieldrin/fodder is 0.027 (mg Dieldrin/kg vegetable per mg Dieldrin/kg soil).

The estimated Dieldrin concentrations in fodder are:

$$C_v = C_{soil\text{-}UCL} \times B_v \text{ Dieldrin/fodder} \times (1-0.12) = 9.18 \times 10\text{-}4 \text{mg/kg}$$
$$C'_v = C_{soil\text{-}max} \times B_v \text{ Dieldrin/fodder} \times (1-0.12) = 1.27 \times 10^{-3} \text{mg/kg}$$

These estimated Dieldrin values in fodder have been compared with regulation Reg. (CE) 396/2005 on MRL of pesticides in food and feed [11]. As regards cereals, the MRL is 0.01 mg/kg. In both cases, the estimated Dieldrin concentration values in fodder show lower values than the maximum residue level.

CONCLUSIONS

The present work describes a possible procedure to evaluate the risk for human health due to the presence of contaminants in agricultural soils; it has been applied in two case studies in contaminated areas included in the list of Italian National Remediation Sites.

The presence of bioaccumulative compounds in soils can potentially affect human health due to the transfer of the pollutants in the foodstuffs grown in the contaminated areas. The index contaminants selected in the case studies, PCBs and Dieldrin, are bioaccumulative compounds that can have a wide range of toxicological effects for human health and procedures are necessary to evaluate their risk.

The methodology described is based on a precautionary principle in both studied cases: in the case of PCBs also carcinogenic effects have been considered while in the case of Dieldrin has been considered also the $C_{soil\text{-}max}$.

Different approaches are used for the evaluation of risk due to the consumption of foodstuffs (*e.g.* the EU methodology) of agricultural soils; in the present work the USEPA approach has been applied in harmonization with the procedures actually used for the risk analysis in potentially contaminated areas for residential, green or industrial use.

In conclusion this study remarks the fact that legislative values for agricultural soils have not yet been derived in Italy and the regulatory limits for the foodstuffs are in force only for a limited number of compounds; then the application of the described procedure or of similar methodologies are considered useful also to cover the mentioned normative gaps.

Conflict of interest statement

There are no potential conflicts of interest or any financial or personal relationships with other people or organizations that could inappropriately bias conduct and findings of this study.

Submitted on invitation.
Accepted on 22 April 2010.

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