PRESENT ENVIRONMENT AND SUSTAINABLE DEVELOPMENT, VOL. 6, no. 2, 2012

HUMAN HEALTH RISK ASSESSMENT OF CONTAMINATED SOILS WITH CARGINOGEN POLLUTANTS

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Key words: carcinogenetic risk, contaminated soils, risk assessment, organic pollutants, inorganic pollutants

Abstract. Human health risk assessment of contaminated soils with carcinogenic pollutants is a multidisciplinary and participative procedure. For this reason, the opportunities of involving the beneficiaries and the entire community in considering human health for environmental assessment have to be improved.

The present paper shows how to use the risk assessment as a method for investigation and evaluation of contaminated soils. The case study used to illustrate the functionality of the risk assessment model is related to an area contaminated mainly with heavy metals. The area is located in Central Romania, close to a specific pollution source – non-ferrous industry. Twenty four soil samples taken from two depth layers of: 0 – 0.2 m, respectively 0.2-0.4 m from an area of 4.000 m^2 were analyzed.

Results of the chemical analysis indicated relatively high concentrations of As, Be, Cd, Cr^{VI}, and Pb, which is matching with the existing pollution levels. Also, in the soil a pollution of organic nature, respectively contamination with polychlorinated biphenyls (PCBs) was identified, its concentration exceeding the intervention threshold established by the Romanian regulation for sensitive uses. Results of the risk assessment revealed a risk factor of contaminated soils of 10^{-4}, with two orders of magnitude above the acceptable risk of 10^{-6} suggested by the World Health Organization (WHO).

Moreover, the contribution of the categories of pollutants (organic/inorganic) to the estimated risk for each exposure pathway considered was illustrated. Therefore, the paper, through the case study

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presented, represents an example of an approach that should be considered by the decision-making factors in approving certain projects, but also by environmental specialists when performing the health risk assessment in relation to certain objectives.
This aspect is especially important as the experience of those performing the human health risk assessments at the planning stage of programs/projects is insufficient and the collaboration with experts from the health sector is uncommon.

Introduction

At international level, the issue of contaminated soil management occurred since the early ‘80s, increasing the concern among the policy makers on this issue. Since there are sufficient data concerning the potential hazard that the contaminated soils pose to the population and environment, in the last years, their management has become a real concern in Romania. In this respect, in 2010, on the Ministry of Environment and Forests website the first version of the *National Strategy for the Management of Contaminated Soils in Romania* (NEPA, 2010) has been published for the public debate.

On the other hand, nowadays, soil pollution has become a widespread infrastructure issue, of different intensity and meanings. The literature data point out that risk-based approaches are vital to allow governments and industries to optimally manage the contaminated sites. But, to determine whether based on the degree of contamination of a site, its use is fully consistent with the chosen use, decisional tools are needed to facilitate the decision process. In this context, there is a need to introduce two very important tools: *Risk Assessment* and *Risk Management*.

The U.S Department of Energy defines the *Risk Assessment* as a process of organizing and analyzing information to determine the probability of a pollutant released in the environment to pose damage to the ecosystem or the exposed population (http://www.lm.doe.gov/Glossary.aspx). The U.S Environmental Protection Agency analyses at the same time with the Risk Assessment the probability for a contaminated area to cause both carcinogenic and non-carcinogenic effects to human health (US EPA, 1992).

The main objective of this paper is to present the methodology of human health risk assessment posed by soils contaminated with carcinogenetic pollutants. The paper presents also the results of the risk assessment as a tool to identify the “hot spots“ in a particular area of interest. The risk assessment represents a very important tool for the decision makers responsible for the management of contaminated sites. Therefore, during the first phase of the research, a physical-chemical description of the area of interest was conducted. The area is located in the Central Romania in close vicinity of a point pollution source – nonferrous industry). The area has a surface of 4.000 m^2. Twelve soil samples were collected from two soil layers: 0-0.2 m, respectively 0.2 – 0.4 m. To identify the level of concentrations of heavy metals in soil (As, Be, Cd, Cr VI, Pb) an atomic absorption spectrophotometer was used, while for determination of the level of concentrations of organic compounds (polychlorinated biphenyls – PCBs) a gas chromatograph coupled with mass spectrometry was used.

Significant results were obtained in terms of risk assessment associated with exposure to a high level of heavy metal concentrations, being well known that the investigated area is one of the most contaminated areas in Europe. Pb and Cd contents have shown the highest level of concentrations for the analyzed soil, inducing a carcinogenic risk above the value indicated by the World Health Organisation (WHO) as being an acceptable risk (10^{-6}, being a cancer case in 1 million persons) (US EPA, 1996).

2.Carcinogenic risk assessement methodology

2.1Generalities. Even if risk has become a central concept in terms of the environmental practice and policy, it has to be underlined that it is not something that can be easily done or defined precisely (Ferguson, C. et al., 1998). Within the National Strategy for the Management of Contaminated Sites in Romania, risk is defined as a potential, predictable hazard, while the Risk Assessment is defined as the determination of the probability for a population or an ecosystem to receive a certain dose of pollutant / polluting substance or to be in contact with the pollutant / exposure (NEPA, 2010). Thus, risk assessment has become, in most cases, an instrument in the undertaking of transparent, rational and well-sustained decisions.

Figure 1 shows a general framework for the management of contaminated sites, having as concept the risk assessment (Swartjes F. A., 2011).

As shown in Fig. 1, the first step in setting the general framework for managing contaminated sites is *Defining of the problem* (contaminated site), or better said its screening. With regard to contaminated sites, Swartjes FA defines four main protection targets to be pursued: 1) human health, 2) soil ecosystem, 4) groundwater, and 5) food safety. The second step to be taken within the general framework of management of contaminated sites is Risk Assessment (or better said the Human Health Risk Assessment). Two types of activities are carried out at this stage: *Exposure assessment* (estimation of the dose) and *Hazard assessment* (estimation of the effect). If in terms of exposure assessment there are mathematical models for its identification, in terms of hazard assessment, the inevitable toxicological properties of the substance must be identified, properties which actually show the intrinsic ability of the chemical element to cause different adverse effects. For example, a chemical compound may be hepatotoxic, mutagen, carcinogen, teratogenic, allergen and so on. This sequence does not inform us if the pollutant shall cause these effects under any circumstances, but simply identifies the main hazards that shall be considered within the assessment to be carried out (EA, 2009). Results of the *Exposure assessment* and *Hazard assessment* lead to the *Risks description.* Thus, the actual assessment of the contaminated site from the point of view of human health risk assessment leads to the intervention, based on the estimated risk and the necessity to take the following step, namely *Risk management.* Risk assessment is carried out to correlate the concentration of pollutant identified within the risk assessment with the human health, ecosystem, underground water or food safety risks. If in terms of exposure assessment there are models for the determination of the exposure, meanwhile, in terms of hazard assessment (of the effect) it is estimated based on the existence or not of threshold limit values (*TLV*) for the contaminant for which the estimation is carried out. For example, the threshold limit value for air is the value of the maximum concentration of vapors or gases from toxic substances to which a person may be exposed for 8 hours per day, for an indefinite period, without any risks to the life or the health of the respective person (Toxicological Reference Value), which is expressed in ppm.

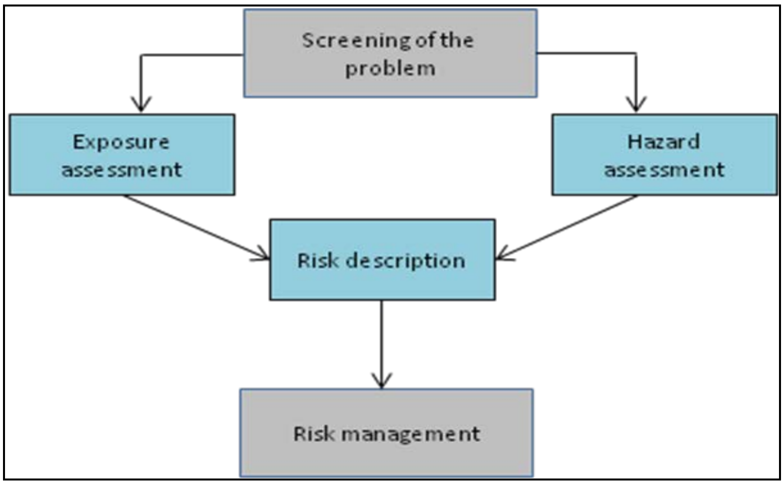

Fig. 1 - General framework for the management of contaminated sites starting from the risk assessment (adaptation after Swartjes F. A., 2011)

2.2. Exposure assessment. There is no exact information in Romania in terms of a specific methodology that should be applied concerning risk assessment. Order no. 184/1997 on the approval of the Procedure for the carrying out of environmental balance sheets, Annex IV (sub-annexes 1 and 2) shows general aspects related to risk assessment (MWFEP, 1997). The following are listed in the

Order: objectives of risk assessment, the types of risk assessment (health assessments and environmental assessment), specific risk elements (chemical risk, carcinogen risk, epidemiological risk, nuclear contamination risk and the risk of occurrence of natural phenomena), identification of the source-way-receiver factors on a contaminated site, the matrix for the analysis of the source-way-receiver ratio. In terms of the qualitative assessment of the risk, Order no. 184/1997 establishes that following factors shall be considered:

(1) *Hazard/source* – specific pollutants identified or supposed to be found on a site, their level of toxicity and their particular effects;

(2) *Exposure pathways* –toxic substances pathway reach the point they have harming effects, either through direct ingestion or direct contact with the skin, or migration through soil, air or water;

(3) *Target/Receiver* – objects on which act the harming effects of certain toxic substances on the site, which may include human beings, animals, plants, water resources and buildings (or their foundations and usage).

Amongst the models for risk assessment presented within the reference strategy, there is also the German model CSOIL (Brand E., et al., 2007). However, it must be underlined that, regardless the exposure assessment model referred to, all these proposed and developed models are based on the exposure assessment models developed by the United States Environment Protection Agency (US EPA, 1992; US EPA, 1997). According to CSOIL, the actuating pathways considered for risk assessment due to exposure to contaminated soil are: *1) related to soil* – ingestion of contaminated particles; dermal contact with contaminated particles (indoor); dermal contact with contaminated particles (outdoor); inhaling of contaminated particles; *2) related to air* – inhaling of contaminants as vapors in enclosed areas (indoor); inhaling of contaminants as vapors (outdoor); *3) related to phytogenous products* – ingestion of contaminants through phytogenous products grown around one's house; *4) related to drinking water* – ingestion of contaminants through drinking water.

2.3. Stages of exposure assessment. This paragraph lists the main steps to be taken during the assessment of the exposure to contaminated sites, as indicated by Swartjes, F. A., 2011. In order to obtain a correct and comprehensive exposure assessment, during the development of the multicriterial model related to risk assessment, inclusion of the most significant aspects considered within the assessments and models proposed by the professional literature was intended.

*2.3.1. Identification of three-phase soil partitioning.*A crucial aspect in exposure assessment (regardless the applied risk estimation model) is the identification of the three-phase partition of the pollutant in soil (solid, liquid and gaseous phases). The distribution coefficients describe the concentration of contaminant in the soil in solid and liquid phase, and liquid and gaseous phase, as presented in equation below (Otte P. F., eta al., 2001):

$$\delta_s \times C_{Tot} = \theta_{pv} \times C_{pw} + \delta_s \times C_s + \theta_a \times C_a \qquad \text{(eq. 1)}$$

where:

C_{Tot} = total concentration of pollutant in the soil ($M/kg_{s.u.}$);

C_{pw} ($M/L_{apă}$), C_s ($M/kg_{s.u.}$) and C_a (M/L_{aer}) = concentrations of pollutant in the soil in liquid, solid, respectively gaseous phase;

δ_s = average soil density (M/L);

θ_w = volumetric water content in the soil (-);

θ_a = volumetric air content in the soil (-).

The concentrations of pollutant in the soil in the mobile phase may be estimated based on the following formula (Otte P. F., et al., 2001):

$$C_{pw} = C_{Tot} / K_d \qquad \text{(eq. 2)}$$

where:

K_d = distribution coefficient between the solid and the liquid phase in the soil (L/M);
H′ = Henry's Law constant (-); distribution of the water/air coefficient.

2.3.2. Transfer of contaminants. The second step in exposure modeling is the prediction of the transfer of contaminants from the mobile phase (for example, from the soil) to a certain environment: air (volatile phase), indoor and outdoor; air (particles); water (underground waters, surface waters, drinking water); dust from houses, phytogenous products: meat, eggs, milk. The accumulation in case of bovine animals may be considered both a pathway of exposure, as well as a way of transfer, this depending on the considered target: assessment of human exposure or bovine exposure. In the end of the estimation of the exposure, the human contact with the pollutants, which may be found in the mentioned compartments, is quantified considering the characteristics of the site, but also the habits of the population (daily routine of the residents).

2.3.3. Main exposure pathways. The majority of the models for the estimation of exposure listed by the professional literature refer to: ● exposure through soil ingestion (oral exposure); ● exposure due to consumption of phytogenous products (oral exposure); ● exposure through indoor air inhaling, excluding inhaling of dust particles (exposure through inhaling). For the listed exposure pathways, the derived intercompartimental links such as the following must be considered: ● links between the concentrations of pollutant in the soil and those from the phytogenous products; ● links between the concentrations of pollutants from the underground waters and soil, and the indoor inhaled air (indoor). Depending on the analyzed context, there are also other exposure pathways listed by the professional literature.

2.4. Exposure assessment. Applying the method of human health risk assessment as per its exposure to a source of pollution leads to the necessity to show in this paragraph aspects related to the quantifying of the exposure. Exposure is the amount of pollutant absorbed by the human body on different ways (inhaling, ingestion, dermal contact) the latter leading the contaminant to the exposed subject (multiple exposure pathways). In other words, risk assessment identifies the cases leading to exposure and determines the absorbed dose by an exposed body or estimates the emission in a particular compartment of the environment (E. Bușa, 2009). The dose absorbed by a subject represents the quantity of a pollutant which may be correlated with the effects induced on human health, this, of course, referring to the body weight unit and the time unit, expressed based on the available toxicological data. If these data refer to doses actually absorbed by the target body, the dose is considered to be equal to the exposure. However, if these refer to the external dose, the dose is determined by multiplying the exposure with a bioavailability factor related to the exposure pathways (for example, pulmonary, gastrointestinal or dermal bioavailability). From a quantitative point of view, the estimation of the exposure is carried out using at the same time information concerning both the level of pollutant in different environment compartments (air, soil, water, food chain), as well as information concerning the duration of the presence of the subjects in different places in which they are object to the exposure and the level of efficiency of the pollutant actually reaching the subject.
The general calculation formula used for the estimation of the exposure may be summarized as follows:

$$I = \frac{C \times CR}{BW} \times \frac{EF \times ED}{AT} \qquad \text{(ec. 3)}$$

where:
I = exposure [mg kg^{-1} zi^{-1}];
C = concentration level of the pollutant in different environment compartments [mg/kg or mg/m^3];

CR = contact factor, meaning the considered quantity from the target environment compartment (air, soil, water, food) in contact with the subject exposed during the time unit [mg/day or m^3/day];
BW = body weight [kg];
EF = frequency of the exposure [day/year];
ED = duration of the exposure [years];
AT = exposure mediation time [years].

In terms of the carcinogenic pollutants, even though the estimation of the risk is the probability of cancer development during the entire lifetime, the exposure is compared to the average lifetime of the exposed subjects, this in spite that, in reality, the exposure period may occur even for shorter periods of time. Thus, the daily average exposure for the entire lifetime is determined (LADD = lifetime average daily dose). For this reason, the AT parameter from the above equation has the value of the average lifetime, considered as being 70 years, i.e. 25,4550 days.

Parameter EF represents the period of the year actually spent by the subject in the contaminated area. The value may be lower than 1 in case it is intended, for example, to limit the period of time to a certain number of days, as in case of the persons who work outside the contaminated area, thus, spending most of their time outside the impact area. In order to illustrate a generally prudent case (preventive) it is correct to consider a unitary value in terms of this parameter.

Parameter ED represents the exposure period (expressed in years) and it may vary between 30 – 70 years. Only in terms of direct exposure (as in the case of exposure through inhaling) this parameter may be allocated the value of 30 years, this period of time being equal with the lifetime of the source of pollution (for example, for a specific source of pollution from the industry – a waste incineration facility). In terms of the direct sources of exposure, the exposure period coincides with the residence period in the contaminated area, and due to prudence reasons, it is recommended to consider a 70 years period, equal to the average lifetime of the subjects. However, actually, during the lifetime of a person, exposure varies depending on age, there being changes in the characteristics of the body, consequently, the air taken in, the quantity and quality of food, the bioavailability of the body, but also one’s habits and life style. Taking into account the aspects listed above, as per the specific parameters of the considered exposure pathways, but also the age of the subjects, the daily doses for a certain pollutant for different exposure pathways, depending on the age, thus depending on the exposure time ED may be determined:

$$I = \frac{\sum_{k=1}^{n} \frac{C \times CR_k \times EF_k}{BW_k}}{AT} \times ED_k \qquad \text{(ec. 4)}$$

where k represents different classes of considered ages. In terms of the age classes, for estimation as accurate as possible of the exposure, the related literature overview shows different intervals of values. However, as a general rule, there are two main groups, adults and children, groups to which are afferent different parameters related to exposure. The exposure for children is considered to be of 9 years, while for adults is considered to be 30 or 70 years.

3. Risk description

Considering the level of pollutants that describe the entire surface of 4.000 m^2 (area of interest mainly contaminated with heavy metals) an assessment of the risk coefficient associated with each contaminant, afferent to each category of pollutants (organic and inorganic) and, also to all pollutants was developed, by summarizing the identified risk coefficients. Several Excel files have been developed for the carrying out of the risk assessment, depending on pollutant type, using tens of parameters identifying the type of pollutant, the exposure, the area where the assessment has been carried out, etc. leading, in the end, to the estimation of the risk coefficient. The risk assessment was mainly based on three documents. First of all, the methodology proposed by the United States Environment Protection Agency in 1992 (US EPA, 1992): Risk Assessment Guidance for Superfund: Volume I – Human Health Evaluation Manual. This document is the starting point for most of the guidelines for risk assessment associated with contaminated soil that were developed by some countries of the European Union (Holland, Germany, Great Britain, France, etc.), Canada and Australia. During the research, defining elements have been also considered, in order to reach a correct estimation of the coefficient associated with soil contamination. Consequently, the CSOIL (Otte, P.F., et al., 2001) Dutch contaminated soil risk assessment model and the guideline of the Ministry of Health from Canada (PQRA, 2004) were studied. Although both approaches (Holland and Canada) are based on the methodology developed by US EPA, 1992, it is extremely interesting to follow different approaches, precisely to finally develop an integrated model considering all aspects and particularities considered by all those who develop guidelines for the development of this type of assessments. To present as accurately as possible the developed risk assessment, the following exposure pathways were considered for the estimation of the risk coefficient: ● exposure through ingestion of contaminated soil; ● exposure through consumption of phytogenous products; ● exposure through dermal contact due to the skin contact with contaminated soil. For the exposure pathways listed above, the following derived intercompartimental links were considered: links between the concentrations of pollutant in the soil and those from the phytogenous products. The soil utility considered for the estimations is the real one, namely the agricultural one.

4. Results and discussions

In terms of exposure assessment, the following have been found for the analyzed metals: ● oral exposure –ingestion of contaminated soil – has the highest contribution from the considered exposure pathways. This is followed by the oral exposure – ingestion of phytogenous products; ● the pollutants for which the quantification of the exposure was developed were accumulated mainly in the surface layer. Thus, the doses with a higher level of absorption by the body are those afferent to the upper soil layer (0 – 0,2 m); ● the increasing tendency of exposures for the considered pollutants has been shown as follows: Be < As < Cd < Cr^{VI} < Pb; ● the exposure carried out through the consumption of vegetal products from the considered area are the main source of exposure for the majority of the heavy metal elements, with the exception of Pb for which the ingestion of soil shows a high level of exposure; ● for the organic pollutants, the ingestion of vegetal products shows the most critical exposure.

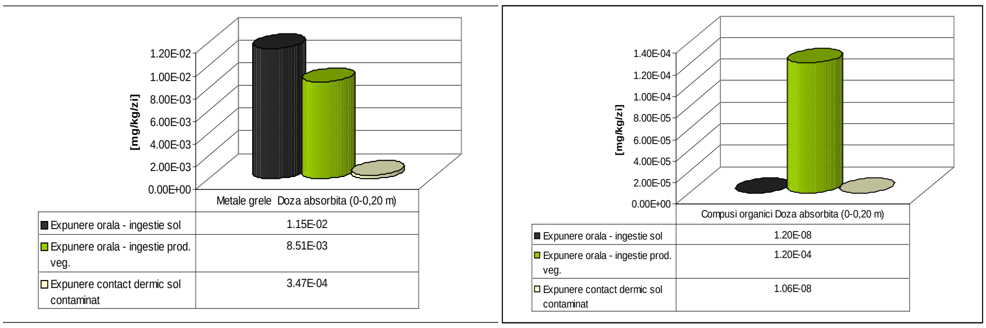

Fig. 2 - Exposure estimation – soil sampling depth (0 – 0,2) m (inorganic and organic contamination)

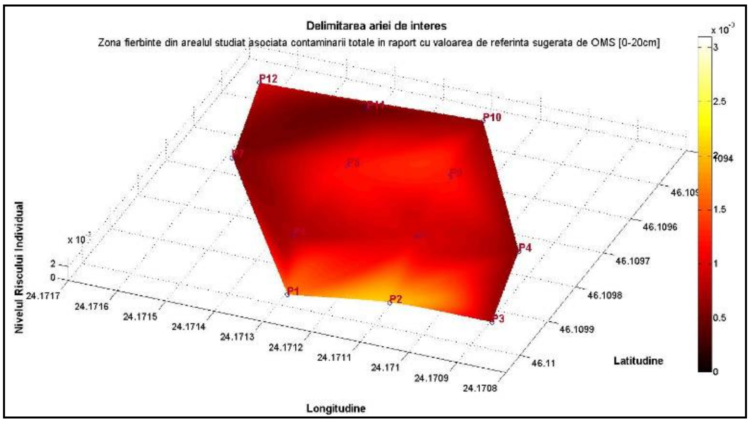

Fig. 3 - Illustrating hot areas from the point of view of distribution of the risk level associated with contamination with heavy metals and organic pollutants (sampling depth 0 – 0.2 m)

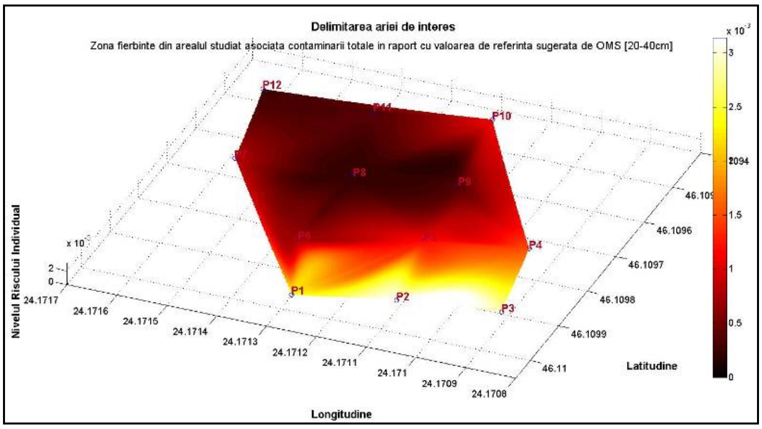

Fig. 4 - Illustrating hot areas from the point of view of distribution of the risk level associated with contamination with heavy metals and organic pollutants (sampling depth 0.2 – 0.4 m)

Thus, in order to identify the “hot spots” from the interest area, risk maps have been developed starting from the level of concentrations of pollutants (organic and inorganic) for each of the 12 sampling points. Based on these levels of concentration, for each pollutant, with the help of an Excel file, the exposures have been estimated, their results being presented briefly in the previous paragraph. With the help of the software application in the MATLAB environment, based on some values of the risk coefficient previously obtained, through extrapolation, and, respectively through the interpolation of the quantified values, the individual risk estimative maps for each category of pollutants were automatically generated. The reference value used for the showing of the “hot spots” was of 10^{-6}, this approach being used for the each of the polluted soil sampling beds: 0 – 0.2 m (Fig. 3) and 0.2 – 0.4 m (Fig. 4).

Conclusions

As a general rule, the human health assessment in correlation with a certain studied objective is missing or it is difficult to identify in the presentations related to human health impact assessment and also in the environmental reports. In most cases, health assessment is at most a qualitative one and not a quantitative one. The results obtained within the present paper lead to the outline of the following conclusions: ● exposure assessment allows the identification of exposure pathways critical from the point of view of the impact on the health of the population residing in an impact area (ingestion of contaminated soil and phytogenous products for pollution with heavy metals and ingestion of phytogenous products for pollution with PCBs in the analyzed case); ● the estimated risk coefficient for the target analyzed area (10^{-4} compared to 10^{-6} the acceptable risk) leads to the necessity to carry out remediation works of the contaminated soil in the respective area; ● risk management should be included in the management of contaminated soils; ● soil decontamination should focus not only on registering under the national limits in terms of soil concentration levels, but more on the reaching of a level of concentrations of pollutants ensuring an acceptable risk ($<10^{-6}$).

Acknowledgement. This paper was developed based on the results obtained within the RECOLAND research project supported by the Operational Programme “Increase of the Economic Competitiveness” POSCCE-A2-O2.1.2.-2009-2, RECOLAND ID519, SMIS-CSNR:11982, Nb. 182/18.06.2010 (2010-2013).

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