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Chapter 20

DETERMINATION OF SCREENING LEVEL FOR SOIL RADIOACTIVE CONTAMINATION

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Abstract: At the present, decisions regarding the clean-up of Brazilian sites contaminated with radioactive isotopes are ressed on a case-by-case basis, since there is no general guidance or recommendation to support actions in the early phases of the problem identification. For chemicals, CETESB - the governmental organization responsible for preventing and controlling environmental pollution in São Paulo State - established quality reference values for prevention and intervention, as the first step to implement a remediation policy based on human health risk assessment. The aim of this study is to develop a methodology for the establishment of target values for radioactive soil contamination as far as possible consistent with and compatible with the approach adopted by CETESB for sites contaminated with chemicals.

The following steps have been addressed in this study: conceptual scenario and model development; codification of the equations in an electronic spreadsheet; selection of proper input values; derivation of the intervention levels for selected radionuclides using Monte Carlo methods. The mathematical model developed was mainly based on the equations used by the U.S. Environmental Protection Agency and by the National Council on Radiation Protection and Measurements for soil screening purposes. Results are presented for selected natural and man-made radioactive isotopes.

Key words: Soil; radioactivity; screening level; target value; contamination.

1. INTRODUCTION

During the 20th century, industrial development was the main cause for the emergence of areas contaminated with harmful substances. During the 1970s, as concerns with environmental issues grew, these areas began to be systematically identified in some countries and clean-up policies implemented. The Superfund program in US is one such example.

In general, soil remediation is an expensive option. Besides, the elapsed time between the identification of the contaminated area and the decision to undertake corrective action typically is too great, due to the complexity of the site-specific studies necessary to a proper characterization of all the associated risks. Despite these problems, governmental agencies from countries such as the Netherlands (VRON, 1988), the USA (U.S.EPA, 1966) and Germany (Bachmann, 2000), among others, have opted to establish target values for soil quality as a first step in the whole process of risk assessment.

In Brazil, the only state to adopt target values for soil contamination – specific to its characteristics – is the State of São Paulo through its governmental agency for pollution control - CETESB. Three levels have been established: (1) quality reference value, indicating the quality level of a soil considered as clean soil, generally associated with the natural concentration of the element of interest; (2) preventing value, above which harmful changes in the soil quality may occur; and (3) intervention value, above which potential risks to the human health exist, considering a generic exposure scenario (CETESB, 2005).

The model used by CETESB to obtain these values was based on that developed by the Netherlands National Institute of Public Health and Environment (VRON, 1994), implemented in the C-SOIL software (Tauw Milieu, 1997).

The list of the elements for which target values are in force includes organic chemicals, volatile organic compounds and heavy metals, all of them potential carcinogens and, therefore, subject to regulatory control. However, it does not include radionuclides. In addition, Brazilian National Nuclear Energy Commission – CNEN –, the federal agency responsible for regulating nuclear activities in the country, has not established target values for radioactive soil contamination either.

This paper describes the methodology proposed to derive intervention levels for radioactive soil contamination in Brazil, as far as possible consistent and compatible with the approach adopted by CETESB for sites contaminated with hazardous chemicals.

2. MATERIALS AND METHODS

Proposed methodology was based mainly on the models developed by the U.S. Environmental Protection Agency (U.S.EPA, 2000a), by the National Council on Radiation Protection and Measurements (NCRP, 1999), and by Companhia de Tecnologia de Saneamento Ambiental (CETESB, 2001). The following exposure pathways were considered: external exposure, inhalation, and ingestion of soil, groundwater, and food.

Values of the input data parameters were chosen using a stochastic approach, according to the best available data for Sao Paulo State; literature data was used otherwise. Intervention levels were derived based on an effective dose to the members of the public of 1 mSv.y^{-1} , taken from the 95th percentile value of the dose distribution.

The annual effective dose for each pathway was calculated according to the following equations:

2.1 External Exposure

$$D_1 = [T_{out} + (T_{in} \times GSF)] \times W_s \times C_s \times ACF \times Df_{ext}$$

D_1 = committed effective dose for external exposure (Sv.yr^{-1})

T_{out} = fraction of time outdoors on contaminated land

T_{in} = fraction of time indoors on contaminated land

GSF = gamma shielding factor

W_s = density correction due to soil moisture

C_s = concentration in soil (Bq.kg^{-1})

ACF = area correction factor

Df_{ext} = external radiation dose factor (Sv.yr^{-1} per Bq.kg^{-1})

2.2 Inhalation

$$Inh = (TSP_{out} \times frs_{out} \times T_{out} \times AV_{out} + TSP_{in} \times frs_{in} \times T_{in} \times AV_{in}) \times C_s \times fr \times fa$$

Inh = inhalation of airborne radionuclides (Bq.yr^{-1})

fr = lung retention factor

fa = absorption factor

TSP = concentration of suspended dust in the air (mg.m^{-3})

frs = fração of soil in the dust

AV = breathing rate ($\text{m}^3.\text{h}^{-1}$)

T = occupation factor (h.d^{-1})

$$D_2 = Inh \times Df_{inh}$$

D_2 = committed effective dose for inhalation of airborne radionuclides (Sv.yr⁻¹)
 Df_{inh} = inhalation dose factor (Sv.Bq⁻¹)

2.3 Ingestion of Soil

$$IngS = AID \times Cs \times fa$$

$IngS$ = ingestion of radionuclides by soil intake (Bq.yr⁻¹)
 AID = annual soil intake (kg.yr⁻¹)

$$D_3 = IngS \times Df_{ing}$$

D_3 = committed effective dose for ingestion of soil (Sv.yr⁻¹)
 Df_{ing} = ingestion dose factor (Sv.Bq⁻¹)

2.4 Ingestion of Groundwater

$$Cpw = \frac{Cs}{\left(Kd + \frac{Vw}{SD}\right)}$$

Cpw = radionuclide concentration in soil pore water (Bq.cm⁻³)
 SD = soil bulk density (g.cm⁻³)
 Vw = water-filled porosity
 Kd = partition coefficient (cm³.g⁻¹)

$$IngA = Qdw \times \frac{Cpw}{DAF} \times fa$$

$IngA$ = ingestion of radionuclides by water intake (Bq.yr⁻¹)
 Qdw = annual water intake (m³.yr⁻¹)
 DAF = dilution/attenuation factor

$$D_4 = InaA \times Df_{ing}$$

D_4 = committed effective dose for ingestion of water (Sv.yr⁻¹)

2.5 Ingestion of Food

2.5.1 Homegrown produce - tubercles

$$Ctb = (Bv_{ib} \times Cs) \times \exp(-\lambda r.th)$$

Ctb = concentration in tubercles (Bq.kg⁻¹)
 Bv_{ib} = root uptake factor (mg.kg⁻¹ wet vegetable / mg.kg⁻¹ dry soil)
 λr = radioactive decay constant (d⁻¹)
 th = time period between harvest and human consumption of the food (d)

2.5.2 Homegrown produce - leaves and fruits

$$Cdp = TSP_{out} \times frs_{out} \times Cs \times DR_{out} \times fi \times [1 - \exp(-\lambda e.te)] \times \frac{1}{Y.\lambda e}$$

Cdp = concentration in vegetables due to foliar deposition (Bq.kg⁻¹)

TSP_{out} = concentration of suspended dust in the outdoor air (kg.m⁻³)

DR_{out} = deposition velocity (m.d⁻¹)

fi = interception factor by edible portion of the vegetable

λe = effective decay constant (d⁻¹)

te = time period that crops are exposed to contamination during the growing season (d)

Y = productivity of the edible portion of the vegetable (kg.m²)

$$Cfl = [(Bv_{fl} \times Cs) + Cdp] \times \exp(-\lambda r.th)$$

Cfl = concentration in leaves and fruits (Bq.kg⁻¹)

Bv_{fl} = root uptake factor (mg.kg⁻¹ wet vegetable / mg.kg⁻¹ dry soil)

$$IngV = (Qtb \times Ctb + Qfl \times Cfl) \times fv \times fa$$

$IngV$ = ingestion of radionuclides by homegrown produce intake (Bq.yr⁻¹)

Qtb = annual intake of tubercles (kg. yr⁻¹)

Qfl = annual intake of leaves and fruits (kg. yr⁻¹)

Ctb = concentration in the tubercles (Bq.kg⁻¹)

Cfl = concentration in leaves and fruits (Bq.kg⁻¹)

fv = fraction of the total consumed vegetables originating from contaminated area

2.5.3 Meat

$$C_f = [(Bv_f \times Cs) + Cdp] \times \exp(-\lambda r.th)$$

C_f = concentration in fodder (Bq.kg⁻¹)

Bv_f = root uptake factor (mg.kg⁻¹ dry vegetable / mg.kg⁻¹ dry soil)

th = time period between harvest and animal consumption of the fodder (d)

$$C_{meat} = C_f \times Q_f \times TQ \times F_{meat}$$

C_{meat} = concentration in meat (Bq.kg⁻¹)

C_f = concentration in fodder (Bq.kg⁻¹)

Q_f = daily animal feed intake (kg.d⁻¹)

TQ = fraction of animal feed originating from contaminated area

F_{meat} = feed to meat transfer factor (d.kg⁻¹)

$$Ing.Meat = Q_{meat} \times C_{meat} \times fv \times fa$$

$Ing.Meat$ = ingestion of radionuclides by meat intake (Bq.yr⁻¹)

Q_{meat} = annual meat intake (kg. yr⁻¹)

C_{meat} = concentration in meat (Bq.kg⁻¹)

fv = fraction of total meat originating from contaminated area

2.5.4 Milk

$$C_{milk} = C_f \times Q_f \times TQ \times F_{milk}$$

C_{milk} = concentration in milk (Bq.L⁻¹)

C_f = concentration in fodder (Bq.kg⁻¹)

Q_f = daily cow milk feed intake (kg.d⁻¹)

TQ = fraction of animal feed originating from contaminated area

F_{milk} = feed to milk transfer factor (d.L⁻¹)

$$Ing.Milk = Q_{milk} \times C_{milk} \times fv \times fa$$

$Ing.Milk$ = ingestion of radionuclides by milk intake (Bq.yr⁻¹)

Q_{milk} = annual milk intake (L. yr⁻¹)

C_{milk} = concentration in milk (Bq.L⁻¹)

fv = fraction of total consumed milk originating from contaminated area

$$IngF = IngV + Ing.Meat + Ing.Milk$$

$$D_5 = IngF \times Df_{ing}$$

$IngF$ = ingestion of radionuclides by total food intake (Bq.yr⁻¹)

D_5 = committed effective dose for ingestion of food (Sv.yr⁻¹)

$$D_{Total} = \sum_{i=1}^5 D_i$$

D_{Total} = total committed effective dose (Sv.yr⁻¹)

Intervention levels were determined for the natural radionuclides ²¹⁰Pb, ²²⁶Ra, ²²⁸Ra, Th-nat and U-nat, and for the man-made radionuclides ⁹⁰Sr, ¹³⁷Cs, ²³⁹Pu e ²⁴¹Am. Three exposure scenarios were considered: agricultural, residential and industrial. Doses to adults only were calculated.

3. RESULTS AND DISCUSSION

Effective doses obtained for each scenario and exposure pathway are presented in Tables 1, 2 and 3. Intervention levels derived in this study are presented in Table 4.

Table 1. Effective dose for agricultural scenario (mSv.ano⁻¹ per Bq.kg⁻¹)

Radionuclide		External Exposure	Inhalation	Soil Ingestion	Grounwater Ingestion	Food Ingestion	Effective Dose
Sr-90	Mean	2,76E-06	1,76E-08	2,04E-06	3,20E-04	7,68E-04	1,09E-03
	Median	2,73E-06	1,41E-08	7,10E-07	5,60E-05	3,57E-04	5,31E-04
	95th %						3,70E-03
Cs-137	Mean	4,31E-04	2,15E-09	7,05E-07	4,06E-07	3,51E-05	4,67E-04
	Median	4,23E-04	1,67E-09	3,33E-07	1,17E-07	2,15E-05	4,59E-04
	95th %						6,55E-04
Pb-210	Mean	5,25E-07	5,75E-07	4,92E-05	8,77E-06	2,17E-04	2,76E-04
	Median	5,05E-07	3,94E-07	1,73E-05	1,87E-06	1,17E-04	1,66E-04
	95th %						8,84E-04

Radionuclide		External Exposure	Inhalation	Soil Ingestion	Grounwater Ingestion	Food Ingestion	Effective Dose
Ra-226	Mean	1,42E-03	1,68E-06	1,67E-05	3,29E-05	8,64E-04	2,33E-03
	Median	1,39E-03	1,28E-06	7,78E-06	3,76E-07	4,90E-04	1,99E-03
	95th %						4,02E-03
Ra-228	Mean	7,62E-04	1,48E-06	6,19E-05	1,44E-04	3,03E-03	4,00E-03
	Median	7,56E-04	8,85E-07	1,72E-05	1,06E-06	1,14E-03	2,02E-03
	95th %						9,69E-03
Th-232	Mean	5,21E-08	1,54E-05	2,00E-05	5,66E-05	2,19E-05	1,14E-04
	Median	5,10E-08	8,90E-06	5,79E-06	3,36E-06	1,04E-05	4,66E-05
	95th %						2,96E-04
U-238	Mean	1,18E-05	1,71E-06	4,04E-06	2,36E-03	8,79E-06	2,38E-03
	Median	1,14E-05	9,87E-07	1,17E-06	6,20E-05	4,08E-06	9,08E-05
	95th %						5,21E-03
Pu-239	Mean	3,33E-08	2,35E-05	1,81E-05	3,39E-05	2,04E-05	9,59E-05
	Median	3,27E-08	1,83E-05	6,37E-06	9,08E-06	1,10E-05	6,35E-05
	95th %						2,32E-04
Am-241	Mean	4,36E-06	2,10E-05	1,48E-05	1,63E-04	1,67E-05	2,20E-04
	Median	4,23E-06	1,44E-05	5,21E-06	4,51E-06	9,01E-06	6,06E-05
	95th %						4,14E-04

Table 2. Effective dose for residential scenario (mSv.ano⁻¹ per Bq.kg⁻¹)

Radionuclide		External Exposure	Inhalation	Soil Ingestion	Food Ingestion	Effective Dose
Sr-90	Mean	1,54E-06	4,77E-08	1,30E-06	3,77E-04	3,80E-04
	Median	1,51E-06	3,59E-08	5,25E-07	1,67E-04	1,72E-04
	95th %					1,30E-03
Cs-137	Mean	2,62E-04	7,16E-09	4,73E-07	1,72E-05	2,79E-04
	Median	2,56E-04	4,30E-09	2,23E-07	1,05E-05	2,73E-04
	95th %					4,08E-04
Pb-210	Mean	2,25E-07	2,16E-06	3,17E-05	1,15E-04	1,49E-04
	Median	2,12E-07	1,04E-06	1,22E-05	6,18E-05	8,90E-05
	95th %					4,61E-04
Ra-226	Mean	8,61E-04	4,84E-06	2,17E-05	4,32E-04	1,32E-03
	Median	8,32E-04	3,30E-06	5,10E-06	2,43E-04	1,16E-03
	95th %					2,24E-03
Ra-228	Mean	4,62E-04	5,35E-06	3,81E-05	1,49E-03	1,99E-03
	Median	4,52E-04	2,22E-06	1,17E-05	5,82E-04	1,08E-03
	95th %					4,64E-03
Th-232	Mean	2,62E-08	6,94E-05	1,25E-05	1,37E-05	9,57E-05
	Median	2,53E-08	2,26E-05	4,20E-06	6,10E-06	4,60E-05
	95th %					2,80E-04
U-238	Mean	5,07E-06	7,69E-06	2,53E-06	4,98E-06	2,03E-05
	Median	4,73E-06	2,50E-06	8,49E-07	2,25E-06	1,33E-05
	95th %					4,78E-05
Pu-239	Mean	1,87E-08	7,80E-05	1,17E-05	1,27E-05	1,02E-04
	Median	1,83E-08	4,73E-05	4,50E-06	6,83E-06	6,87E-05
	95th %					2,67E-04
Am-241	Mean	2,20E-06	7,90E-05	9,54E-06	1,06E-05	1,01E-04
	Median	2,10E-06	3,81E-05	3,68E-06	5,65E-06	5,84E-05
	95th %					2,74E-04

Table 3. Effective dose for industrial scenario (mSv.ano⁻¹ per Bq.kg⁻¹)

Radionuclide		External Exposure	Inhalation	Soil Ingestion	Food Ingestion	Effective Dose
Sr-90	Mean	8,79E-07	3,65E-07	6,48E-07	7,44E-05	7,63E-05
	Median	8,56E-07	2,88E-07	2,41E-07	3,59E-05	3,74E-05
	95th %					2,58E-04

Radionuclide		External Exposure	Inhalation	Soil Ingestion	Food Ingestion	Effective Dose
Cs-137	Mean	1,44E-04	4,38E-08	2,25E-07	3,76E-06	1,48E-04
	Median	1,40E-04	3,47E-08	1,08E-07	2,30E-06	1,43E-04
	95th %					2,17E-04
Pb-210	Mean	1,41E-07	1,17E-05	1,55E-05	4,96E-05	7,70E-05
	Median	1,32E-07	8,48E-06	5,85E-06	1,84E-05	4,15E-05
	95th %					2,10E-04
Ra-226	Mean	4,77E-04	4,24E-05	5,85E-06	9,24E-05	6,18E-04
	Median	4,61E-04	2,55E-05	2,35E-06	5,44E-05	5,77E-04
	95th %					9,48E-04
Ra-228	Mean	2,55E-04	3,17E-05	1,98E-05	3,13E-04	6,20E-04
	Median	2,49E-04	1,91E-05	5,89E-06	1,29E-04	4,35E-04
	95th %					1,28E-03
Th-232	Mean	1,54E-08	3,15E-04	6,24E-06	1,43E-05	3,36E-04
	Median	1,47E-08	1,86E-04	1,90E-06	2,55E-06	1,97E-04
	95th %					9,96E-04
U-238	Mean	3,18E-06	3,49E-05	1,26E-06	3,35E-06	4,27E-05
	Median	2,94E-06	2,06E-05	3,84E-07	7,77E-07	2,70E-05
	95th %					1,13E-04
Pu-239	Mean	1,06E-08	4,79E-04	5,71E-06	1,20E-05	4,97E-04
	Median	1,02E-08	3,80E-04	2,15E-06	2,75E-06	3,94E-04
	95th %					1,18E-03
Am-241	Mean	1,29E-06	4,29E-04	4,68E-06	9,92E-06	4,44E-04
	Median	1,22E-06	3,10E-04	1,76E-06	2,42E-06	3,21E-04
	95th %					1,16E-03

Table 4. Intervention Values for Each Scenario

Radionuclide	Intervention value (Bq.kg ⁻¹)		
	Agricultural	Residential	Industrial
⁹⁰ Sr	270	770	3880
¹³⁷ Cs	1530	2450	4610
²¹⁰ Pb	1130	2170	4760
²²⁶ Ra	250	450	1055
²²⁸ Ra	100	220	780
²³² Th	3380	3570	1000
²³⁸ U	190	20920	8850
²³⁹ Pu	4310	3740	850
²⁴¹ Am	2420	3650	860

According to Hiromoto et al. (Hiromoto, 2006), geometric mean background activity concentration values in soils of Sao Paulo State for ¹³⁷Cs, ²¹⁰Pb, ²²⁶Ra, ²²⁸Ra, Th-nat and U-nat are, respectively, 1.9, 46, 17.1, 27.8, 30 and 93 Bq.kg⁻¹ dry weight. Therefore, these results show that intervention values are orders of magnitude higher than background levels for most of these radionuclides, except for ²²⁸Ra and U-nat. In these two cases, intervention values are somewhat closer to the background levels, showing that site-specific data is necessary, in some circumstances, even for screening purposes.

It was observed that, as a general rule, external exposure is the critical pathway for gamma emitters, vegetable ingestion is the critical pathway for beta emitters, and inhalation of suspended particulates, is the critical pathway for alpha emitters. This behavior is more clearly observed in the industrial scenario, where the concentration of dust in the air, that is a user input data, is much higher than in the residential and agricultural scenarios.

4. CONCLUSION

The intervention levels obtained in this study agree with those reported in the NCRP-129 (NCRP, 1999), considering the differences between both methodologies and input data. They also, show that

the model is robust and compatible with the methodology adopted by CETESB for the risk assessment of chemicals.

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