Environmental impact of radioactivity in waste from the coal and aluminium industries in western Balkan countries

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EC INCO-509214 INTAILRISK project partners: Assessment of Environmental Risk of Radioactively Contaminated Industrial Tailings. Contact addresses on http://www.ibes.be/intailrisk

Abstract. This paper deals with industrial tailings resulting from the use of radioactive coal and bauxite for assessing the impact on the population and the environment in the western Balkan countries (WBC). It considers the direct hazard resulting from the wastes for their immediate neighbourhood and the radionuclide dispersal in the environment through surface and groundwater. The selected test sites have been investigated by different methods assessing the presence and type of radionuclides in the primary and waste products, analyzing and identifying the pathways for dispersion of radionuclides in the waste surroundings, and defining the impact of the waste on the ecosystem. The process of leachability and fractionation of the different radionuclides has been studied, too. The transport of radionuclides in groundwater has been studied by 3D groundwater flow and solute transport modelling. The following parameters have been assessed (1) gamma dose-rate levels (2) radon in soil gas (3) radon exhalation (4) indoor and outdoor radon (5) radionuclide activity in soil and in waste material (6) radionuclides in surface water and groundwater (7) radionuclides in biota. Several case studies highlight the transfer of the radionuclides to plants and animal consumption products. The radionuclide concentrations in the waste and in the surroundings range over three orders of magnitude. Radionuclide concentrations in groundwater surrounding the waste are low and have a lower variability than in the wastes. Radon concentrations on the tailings are increased with respect to the surroundings. The transfer factors in the soil–plant–animal system indicate low bioavailability of investigated radionuclides. A preliminary dose assessment shows that the highest contribution to dose is from external and radon exposure. On the basis of the results obtained by using transport model simulation and considering the high Kd values and low concentration of radium, radionuclide transport in groundwater is slow and limited to a restricted area around the tailings sites.

1. Introduction

The EC (6th Framework) International Cooperation (INCO) project 509214 *Assessment of environmental risk of radioactively contaminated industrial tailings (INTAILRISK)* investigates waste containing NORM material from coal burning power plants (CBPP) and the aluminium industry in western Balkan countries (WBC). The objectives of the project are to assess the possible hazards to the public and the environment emerging from radionuclides in the studied wastes (tailings, derived products). The project investigates the risk from the waste itself, the dispersion of radionuclides from the waste to the environment, and the possible effects from the secondary (derived) products. In the frame of the project, the possible hazards for radionuclide leaching from the waste and contaminating groundwater, surface water, soil and agricultural products have been studied at different test sites, selected on the basis of their specific context (age, remediation actions, climate, subsurface characteristics, distance to living areas etc.). The test sites in the different participating countries have been characterized in detail in terms of radionuclide type, distribution and dispersion. The current paper focuses on the results of the uptake of radionuclides in biota and on the results of contaminant transport model.

2. Methodology

2.1. Site characterization

The test sites, including the tailings ponds and their surroundings up to nearby living areas, and sometimes the industrial facilities and the raw materials, have been characterized by (1) gamma dose-rate measurements (2) radon in soil gas (3) radon exhalation (4) indoor and outdoor radon

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concentration (5) radionuclide activity concentration in soil and waste material (6) radionuclides in surface water and groundwater (7) radionuclide concentrations in biota. A description of the different studied test sites and the applied methodology can be found on the project website www.ibes.be/intailrisk.

2.2. Leachability study

Speciation studies of radionuclides and trace elements involve sequential or parallel leaching experiments using selective reagents to extract radionuclides associated with particular phases of the investigated materials. Literature data indicate that the parallel extraction method has been proved useful in investigating the main differences in solid speciation of radionuclides in agricultural soils, particularly when taken in junction with the results of ultra-filtration study.

The most frequently used methods for speciation studies of natural radionuclides (^{238}U and ^{232}Th series) are Tessier's method, Schultz's method as well as the standardized 3-step sequential extraction technique developed by a European working group coordinated and supported by the Bureau Commun de Reference, known as the BCR technique. Previous studies have not provided statistically significant quantification of leachability and bioavailability of radionuclides, which can be widely used in environmental assessment models. Therefore, some of the participating laboratories have developed their own methods based on the above mentioned methods. The results obtained by those techniques have been compared to the BCR method. Despite that fact, it was necessary to perform a leachability study for the purpose of improving the knowledge about capability of radionuclides for migration processes.

2.3. Contaminant transport modelling

3D reactive and non-reactive transport models of contaminants in groundwater have been studied using the MODFLOW and MT3D codes. MODFLOW solves the partial differential 3D flow equation by using a finite differences method:

$$\frac{\partial}{\partial x_i}\left(K_{ij}\frac{\partial h}{\partial x_j}\right) = S_s\frac{\partial h}{\partial t} + Q$$

where:

S_s is the specific storage coefficient

h is the hydraulic head

K_{ij} is the hydraulic conductivity

Q is the recharge

i,j are the 1, 2, 3 main directions

A numerical model implemented in Visual MODFLOW requires:

1. The horizontal and vertical discreetization of the domain in cells. The solution is assigned to the node located at the centre of the cells;
2. The type of simulation (steady state or transient) and initial conditions;
3. The main hydrogeological parameters for each cell: hydraulic conductivity, porosity, storage coefficient;
4. The boundary conditions (i.e. assigned hydraulic head: h=cost, assigned flux to a cell: q =cost for pumping wells, recharge, inactive cells indicating no flux condition; variable flux that depends on the hydraulic head in the cell).
5. The model calibration that uses the parameters estimation procedure (PEST).

$$\underbrace{\frac{\partial c}{\partial t}}_{I} + \underbrace{\frac{(1-n)}{n}\rho_s \frac{\partial c_a}{\partial t}}_{IV} = \underbrace{-\frac{\partial}{\partial x_i}(v_i c)}_{II} + \underbrace{\frac{\partial}{\partial x_i}\left(D_{ij}\frac{\partial c}{\partial x_i}\right)}_{III} - \underbrace{\lambda c}_{V} - \underbrace{\sum Q c_{in}}_{VI}$$

MT3D solves in 3D the advection-dispersion equation of a contaminant in the aquifer. Different terms indicate: I. initial concentration, II. advection, III. dispersion, IV. adsorption, V.decay, VI. recharge or loss. MT3D automatically uses MODFLOW results to solve the transport equation. It needs the horizontal and vertical dispersivity coefficients as input parameters, the definition of the initial concentration (at time t = 0); the definition of the boundary conditions (presence of source with constant concentrations, inactive cells and cells with variable concentrations). The model was constructed using geological, hydrogeological and hydrochemical data from wells located in the source area (TENT B thermo-powerplant, Obrenovac, Serbia and Montenegro) as well as its surroundings. The transport model was implemented by using reactive, soluble sulphates in combination with radium as a passive contaminant.

3. Results

3.1. Dispersion of radionuclides around the waste

As a general observation, the distribution of radionuclides in soil around the investigated tailings is limited to the close vicinity of the tailings, and does not extend to distances larger than about 100 m. Fig. 1 shows an example of gamma dose rate distribution around the tailings area of the Kakanj CBPP (Bosnia).

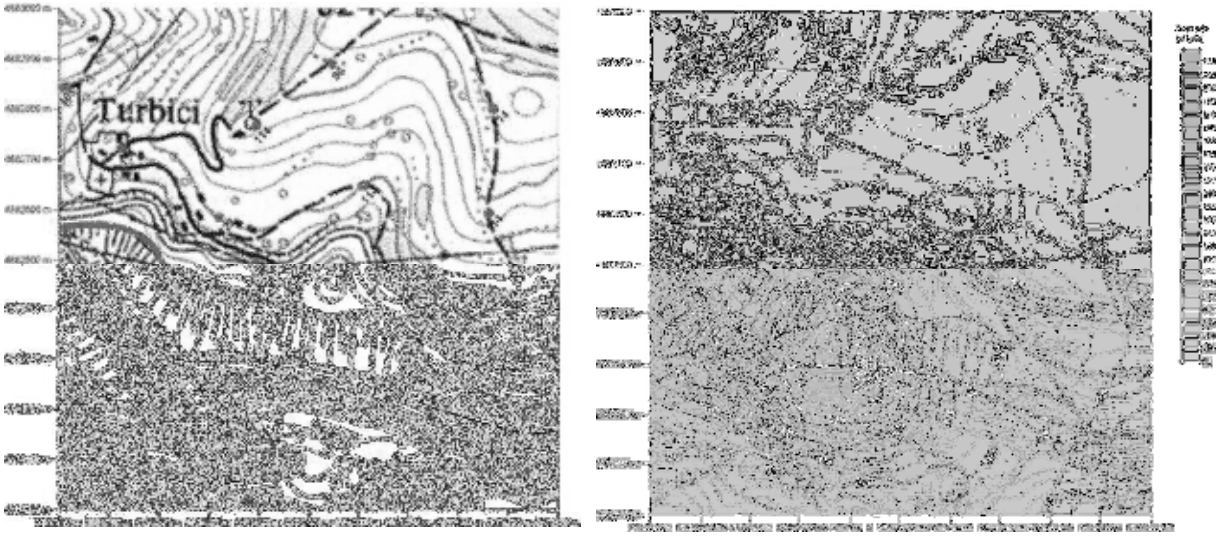

FIG. 1. Dose rate map of the Kakanj CBPP depository (Bosnia). Tailings area outlined in grey on the left side image.

Another example (Fig. 2) shows that the radon exhalation rate remains smal, and is only increased in the tailings area.

3.2. Radionuclide leaching to the groundwater

At the test site Kansk Achinsk of the Siberian research group, joined to the project, the radionuclide distribution in depth soil profiles highlight disequilibrium between ^{238}U and Ra/Pb, with relative depletion of the former radionuclide. This indicates the possible migration of U to deeper levels or leaching to the groundwater. Fig. 3 illustrates this type of disequilibrium on the test-site of Kansk Achinsk in Siberia.

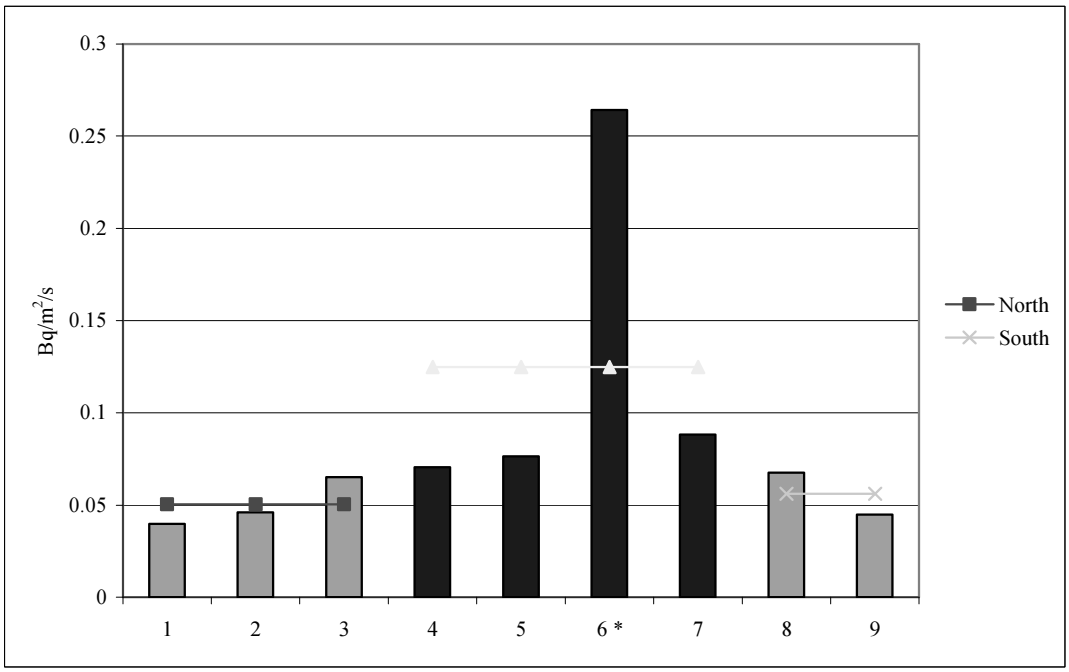

FIG. 2. Radon exhalation rate on and around the waste depository of Kocevje coal mine (Slovenia). Black bars on the depository, grey bars outside the depository.

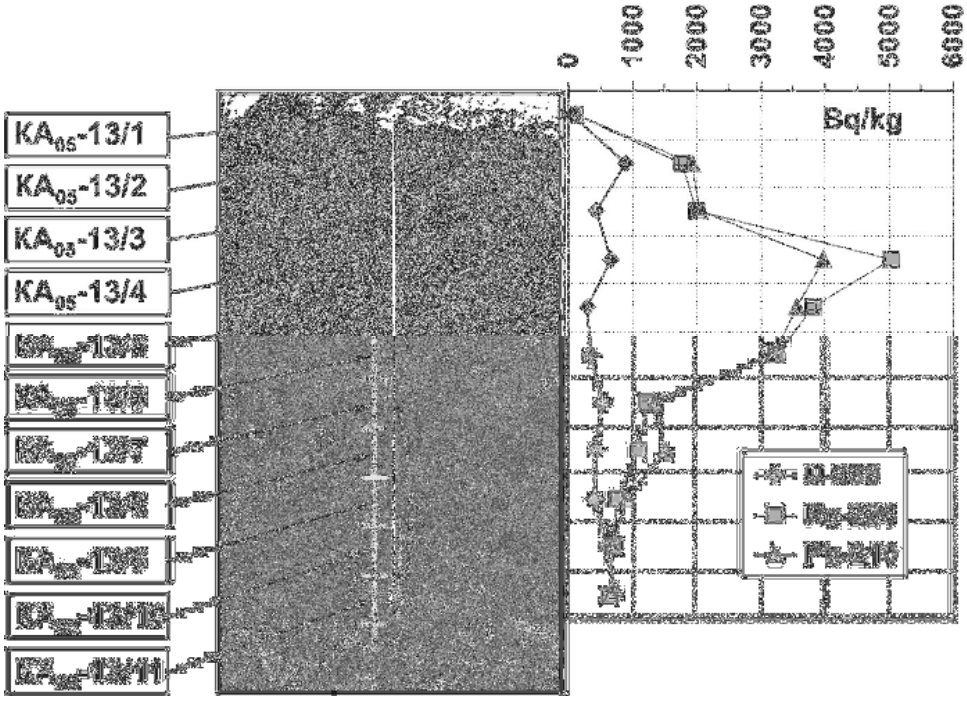

FIG. 3. Radionuclide disequilibrium in soil profiles at the test-site of Kansk Achinsk (Siberia).

3.3. Radionuclide uptake in biota (agricultural products)

Sampling was performed at two different points in Bosnia and Herzegovina. Point A was sampled in the vicinity of the coal burning power plant Kakanj in the central part of Bosnia and Herzegovina, while the point B placed in the vicinity of the coal mine Tusnica-Livno in the south-western part of

Bosnia and Herzegovina. The main soil parameters at points A (coal burning power plant Kakanj) and B (coal mine Tusnica-Livno) are shown in Table 1.

TABLE 1. SOIL PARAMETERS AT INVESTIGATED POINTS

	pH	Humus (%)	Humidity (%)	Minerals (%)	Organic matter %	Type of soil
Point A, Kakanj	7.63	3.9	2.16	89.54	8.3	Loamy soil
Point B, Tusnica-Livno	7.77	6.1	4.87	76.67	18.46	Loamy soil

Radioactivity concentrations in analyzed agricultural products (Fig. 2) did not indicate any significant contamination of the agricultural products. Transfer factors obtained for point A were generally higher than those recorded at point B despite the fact that the radioactivity concentrations of ^{238}U and ^{226}Ra in the soil at point B were significantly higher owing to the use of coal ash for fertilization of the agricultural land.

TABLE 2. RADIOACTIVITY CONCENTRATION AND TRANSFER FACTOR (Dry weight)

		^{238}U		^{232}Th		^{226}Ra	
	Point	Activity concentration (Bq/kg)	Transfer factor (kg plant per kg soil)	Activity concentration (Bq/kg)	Transfer factor (kg plant per kg soil)	Activity concentration (Bq/kg)	Transfer factor (kg plant per kg soil)
Soil 0–15 cm	A	41.12 ± 4.40		31.39 ± 1.3		26.60 ± 1.11	
	B	141.50 ± 10.80		39.53 ± 1.3		196.90 ± 1.90	
Grass	A	2.09 ± 0.41	0.0508 ± 0.0113	0.36 ± 0.05	0.0114 ± 0.0017	3.20 ± 0.31	0.1204 ± 0.0125
	B	4.39 ± 0.71	0.0310 ± 0.0055	0.33 ± 0.05	0.0084 ± 0.0012	7.17 ± 0.66	0.0364 ± 0.0034
Hay	A	1.53 ± 0.22	0.0371 ± 0.0067	0.19 ± 0.02	0.0061 ± 0.0008	1.93 ± 0.18	0.0098 ± 0.0009
	B	2.62 ± 0.34	0.0185 ± 0.0028	0.21 ± 0.02	0.0052 ± 0.0006	4.86 ± 0.41	0.0247 ± 0.0021
Corn stalk	A	0.82 ± 0.15	0.0200 ± 0.0041	0.13 ± 0.02	0.0040 ± 0.0008	1.19 ± 0.12	0.0449 ± 0.005
	B	1.58 ± 0.28	0.0112 ± 0.0021	0.14 ± 0.03	0.0035 ± 0.0007	1.80 ± 0.19	0.0091 ± 0.0009
Corn	A	0.10 ± 0.04	0.0025 ± 0.0010	< 0.02		0.18 ± 0.02	0.0009 ± 0.0001
	B	0.13 ± 0.05	0.0009 ± 0.0003	0.05 ± 0.02	0.0012 ± 0.0004	0.27 ± 0.03	0.0014 ± 0.0001
Bean	A	0.07 ± 0.02	0.0017 ± 0.0006	0.04 ± 0.01	0.0011 ± 0.0004	0.08 ± 0.01	0.0031 ± 0.0005
	B	0.20 ± 0.06	0.0014 ± 0.0004	0.05 ± 0.01	0.0012 ± 0.0003	0.32 ± 0.04	0.0016 ± 0.0002
Potato	A	0.42 ± 0.14	0.0103 ± 0.0036	0.28 ± 0.05	0.0090 ± 0.0015	0.19 ± 0.05	0.0071 ± 0.0018
	B	0.67 ± 0.20	0.0047 ± 0.0014	0.27 ± 0.04	0.0070 ± 0.0010	0.67 ± 0.08	0.0034 ± 0.0004
Turnip	A	0.29 ± 0.15	0.0070 ± 0.0036	< 0.02		0.36 ± 0.07	0.0138 ± 0.0028
	B	0.66 ± 0.25	0.0046 ± 0.0018	< 0.02		1.07 ± 0.16	0.0054 ± 0.0008

The differences between points A and B could be explained by the fact that point A was in the vicinity of a coal burning power plant and was therefore affected by the fly ash from the facility as well as by ash from the waste disposal site (dry type). As a result, higher percentages of ash for unwashed samples of grass, hay and corn stalk from point A were recorded compared with the same samples from point B. That finding indicated the possibility of surface contamination of the green vegetables in the case of a significantly increased radioactivity concentration in the fly ash from the coal burning power plants. Additionally, the soil type and soil parameters at point B (loamy soil, high content of organic matter 18.5 %) indicated a lower migration potential of natural radionuclides. Generally it is reported that the uranium transfer factor decreases from sandy to clayey soils [1, 2]. The presence of organic matter generally decreases the uranium transfer factor [1, 3, 4].

The transfer factors obtained are in good agreement with literature data, taking into consideration the fact that transfer factors usually vary over a wide range as well as that the transfer factors of natural radionuclides for agricultural products are generally low. There are several reports that, among

agricultural products, leafy vegetables generally show higher transfer factors, followed by root, fruit and grain crops [1, 5, 6].

According to the transfer factors obtained in the soil-plant system, the transfer of ^{238}U, ^{232}Th and ^{226}Ra to biota is limited. The dose by ingestion of the agricultural products, therefore, would not significantly contribute to the total dose in the case of acceptable levels of these radionuclides in the waste and soil. The risk from ingestion of the agricultural products would be higher in the case of significantly increased levels of natural radionuclides in waste as well as in areas with a high natural background.

3.4. Radionuclide contaminant transport model

Considering the fly ash deposits (6 km^2) in the TENT B area as the possible source of groundwater contamination, the radium content (about 0.8 Bq/L) in surface waters and groundwater was treated as a passive contaminant (i.e. non-reactive); otherwise, the sulphate content (400 mg/L) measured in the wells of the deposits was treated as a reactive contaminant (i.e. soluble compound with natural retardation). In order to model the worst scenario for the radium transport, only the advection term (i.e. transport of the contaminant at the same velocity as the groundwater flow) and the dispersion term of the advection–dispersion equation of a contaminant transport in the aquifer was considered. In the case of sulphates, a further transport term was included: the retardation (i.e. transport delay caused by the soil/water partitioning of the contaminant). The constant partition coefficient, Kd, a function of the contaminant, was tuned in the range 0–1000 mg/L.

The maps shown in Figs 4 and 5 show the results of the simulation of radium particle migration and of sulphates from the deposit considering the advection and dispersion terms of the equation, as well as the effect of an increasing Kd on the plume extent for the sulphate. The simulation time was 10 years. For the studied test-site, *in casu* tailings of 'TENT B' CBPP in Serbia, the simulation results show the absence of any significant Ra migration around the confined depository.

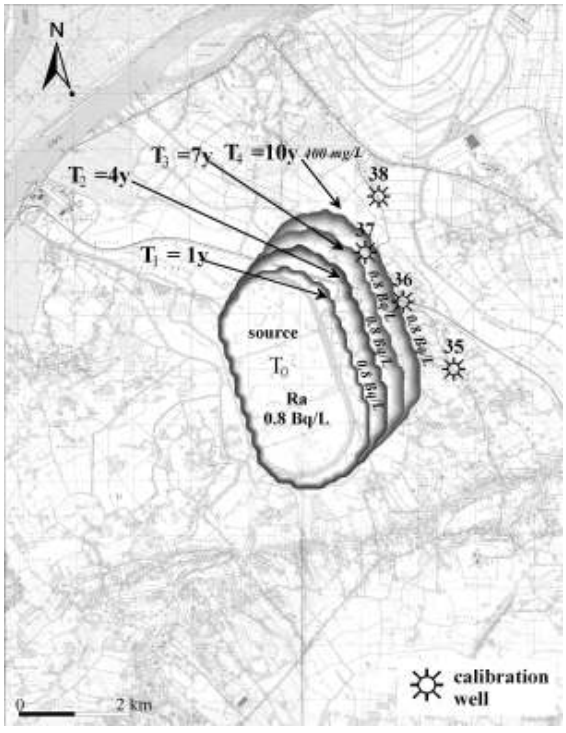

FIG. 4. Ra contaminant transport modelling results. Isolines represent the simulation of the source concentrations at different times up to 10 years. The symbols indicates some sampled wells used as calibration points.

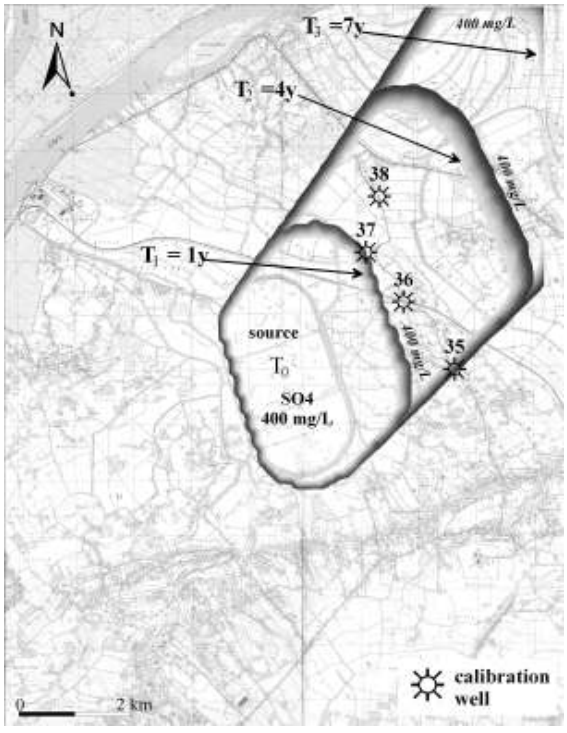

FIG. 5. Sulphate transport modelling results considered as soluble compound. Isolines represent the simulation of the source concentrations at different times up to 10 years. The symbols indicates some sampled wells used as calibration points.

Furthermore, in order to highlight how the plume concentration changes over time down-gradient from the initial source area, a monitoring well with a known concentration of sulphates is added. This monitoring well is used to assess the plume concentration at this point by displaying a concentration vs time breakthrough curve. Considering for sulphates a value of Kd = 0.2 mL/g (the most realistic situation), after 10 years the concentration at the monitoring wells is of the same order of magnitude of the measured concentration (Fig. 6).

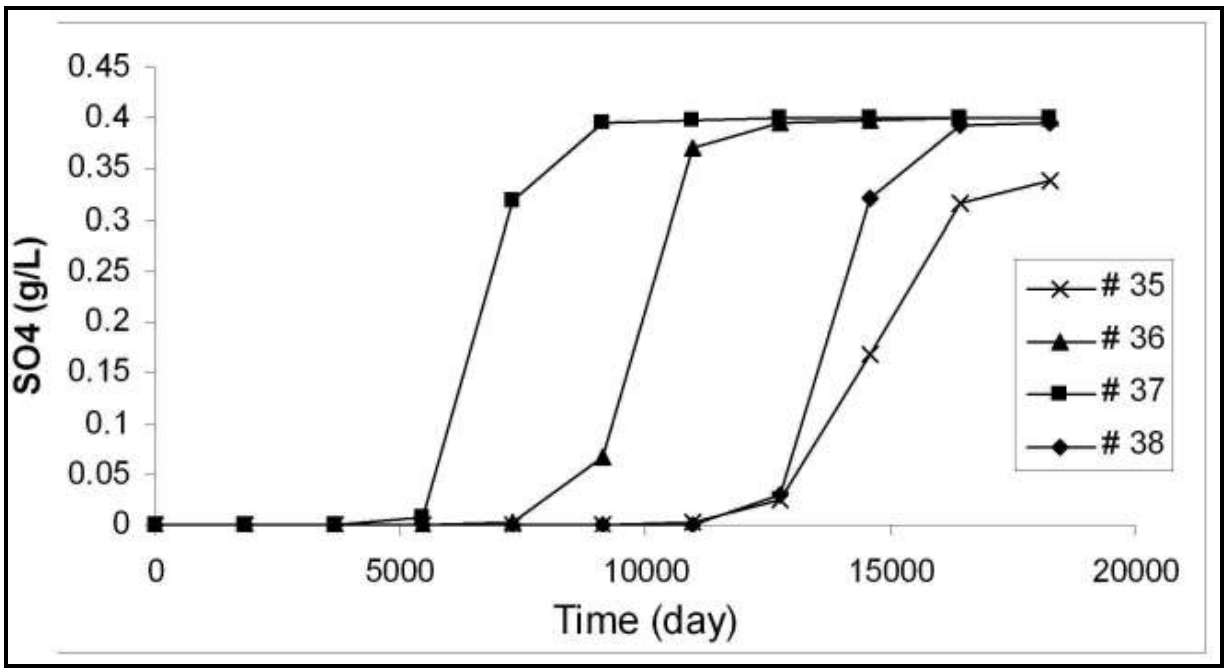

FIG 6. Calibration curves of the sulphates (Kd = 0.2 mL/g) calculated for some sampled wells located in the western side of the studied area. In general, the source concentration is reached in the different wells in the range between 8000-10 000 d.

4. Discussion and conclusions

The impact of radionuclides from tailings from the coal and aluminium industry studied on several relevant and specific test sites by different teams of the project consortium are qualitatively summarized in Table 3.

TABLE 3. OVERVIEW OF PARAMETERS STUDIED IN AND AROUND CBPP AND ALUMINIUM INDUSTRIAL TAILINGS AND THEIR GENERALIZED QUALITATIVE IMPACT

Studied parameter	Generalized observed impact
Gamma exposure	Limited to the close vicinity of the tailings
Radon in soil	Limited to the close vicinity of the tailings
Radon exhalation	Limited to the close vicinity of the tailings
Radon in indoor and outdoor air	Sometimes increased in settlements close to tailings
Radionuclides in soil	Limited to the close vicinity of the tailings
Radionuclides in water	Low to moderate concentrations
Radionuclides in biota	Low to moderate concentrations

It can be generally stated that the surface contamination by radionuclides in the studied sites remains limited to the close periphery of the waste sites themselves.

Groundwater downstream of the waste facilities is not, or only weakly, affected by the radionuclides.

On the basis of the recorded radioactivity concentrations in agricultural products and calculated transfer factors in the soil–plant system, it can be stated that bioavailability of the observed

radionuclides is generally low, and hence the radiation risk coming from the ingestion of agricultural products is limited.

The main (but limited) contribution to the dose received by the surrounding population comes from (indoor) radon.

Contaminant transport modelling indicates a limited radionuclide contamination and dispersion in groundwater. Greater hazards arise from heavy metals and organic pollutants.

ACKNOWLEDGEMENT

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