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Assessment of the mineral industry NORM/TENORM disposal in hazardous landfills

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Abstract

The main objective of this paper is to describe the assessment methodology utilised in Brazil, to foresee the performance of industrial landfills to disposal solid wastes containing natural radionuclides arising from milling and metallurgical installations that process ores containing NORM. An integrated methodology is utilized and issues as risk, exposure pathways and the plausible scenarios in which the contaminant can migrate and reach the environment and human beings are addressed. A specific example of the procedure is described and results are presented for actual situations. The model consists of an engineered depository constructed of earthen materials which minimise costs and maintain integrity over long-term. In order to define the landfill characteristics and the potential consequences to the environment, an impact analysis is carried out, considering the engineering aspects of the waste deposit and the exposure pathways by which the contaminant can migrate and reach the environment and human beings. Analytical solutions are used in the computer program in order to obtain fast results.

Keywords: Solid waste disposal; Industrial landfill; Natural radioactivity series; TENORM; Mining and milling

1. Introduction

There are several circumstances in which materials containing natural radionuclides are recovered and processed that may lead to enhanced concentrations in the final products or wastes, in such a way that the radiation exposure results in significant doses to the public.

The exposures generally included in the category of enhanced exposures are those arising from the mineral processing industries and from fossil fuel combustion. Industry uses many different types of raw materials that contain naturally occurring radioactive materials, sometimes abbreviated as NORM. The raw materials are mined, transported, and processed for further use. During the process to obtain the product, wastes and by-products containing enhanced natural radioactivity – the TENORM material – are generated. The natural radionuclides present in those raw materials or wastes are those of three naturally occurring series: uranium series (U-238), actinium series (U-235) and thorium series (Th-232) [1,2]. These wastes are produced in very large volumes with relatively low specific activities and must be disposed in a way that ensures they remain sufficiently isolated as long as necessary to protect the human health.

Different industrial processes use a mineral feed containing the NORM isotopes, where the parent nuclides (U-238 and Th-232) are more or less in equilibrium with their progeny [3]. For example, in the pyrometallurgy to recover Nb/Ta, all radioactive species become more concentrated in the waste, except the radioactive lead that is volatilized during the process; similarly in the conversion of phosphate rock into fertilizer, using hydrometallurgy process, the by-product phosphogypsum receives most of the radium and its progeny.

The main objective of the paper is to present a methodology for the long-term prediction of the environmental impact of landfills used for the disposal of these solid radioactive wastes that result from the mineral industry. The scenario assumed is that the industrial landfill, constructed of earthen materials is placed inside the installation, in an engineered structure above an aquifer. The earthen materials maintain the deposit integrity through large geologic times (thousand of years), and have a low cost of construction for the depository. The pathway analysis is used to evaluate the facility design for disposal

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of NORM/TENORM. All the main phenomena involved in the waste migration as well as the main pathways are taken into consideration, and an example of the procedure is described using the slag produced from typical Brazilian installations.

2. Overview of metal extraction processes and wastes generation

It is generally believed that the level of NORM found in ores depends more on geologic formation or region rather than on the particular type of mineral being mined. These ores often contain many different minerals, and the radionuclides contained in a certain type of ore or mining operation or its wastes will not be representative of other mines or waste types. But there are some conventional ores like zircon, niobium/tantalum, phosphates and gold that, due to their geological or mineralogical characteristics, have a strong affinity with uranium and or thorium. Also, some processes associated with metal extraction appear to concentrate certain radionuclides and enhance their environmental mobility. In Brazil's extensive territory mining and processing mineral ores with significant amount of uranium and or thorium associated (TENORM) can be classified in the following groups: (1) the Nb/Ta carbonatitic and pegmatitic deposits; (2) the Sn, Ta/Nb, Zr deposits related to intrusive granitic bodies; (3) the Zr, Ti, monazite deposits in sand ores; (4) the phosphate deposits; (5) the Cu deposits in the Carajás mineral province.

The mining, milling and metallurgical installations generally process ores of low grade to recover and produce refined metals of high purity. The metals must then be concentrated and purified requiring a great deal of physical and chemical treatments to separate and liberate them from wastes. These treatments involve operations of mineral beneficiation and extraction by metallurgy. During the mineral beneficiation, part of the rock is discarded as valueless material, while the concentrate of the mineral compound goes on to extraction to produce the metal, usually by pyrometallurgy or hydrometallurgy.

Pyrometallurgy involves operations where furnace treatments at high temperatures are used to separate the metal values from the considerable amount of waste rock still present in the beneficiated concentrate. This waste product is most often removed as slag, which is usually discarded for disposal.

Hydrometallurgical extraction is the leaching of the metal value taken out of an ore or concentrate in an aqueous solvent. This material leaves in the leach solution that must then be purified in order to produce high grade concentrates. The purifying operations of leach solution by solvent extraction, ion exchange, adsorption, crystallization and ionic precipitation often generate barren solutions, which must be treated to neutralize and precipitate the contaminants. The solid precipitated is also a residue to be discarded. An example of hydrometallurgy can be found in fertilizer production, where the phosphate rock is usually converted into phosphoric acid by sulfuric acid reaction. In this process, the secular equilibrium is not maintained and the U and Th daughters remain in the produced waste, named phosphogypsum.

3. Methodology

The aim of a risk assessment for a NORM/TENORM waste disposal facility is to demonstrate compliance with the safety requirements, related to human exposure and the environment. These results are used to judge the design's ability to meet the radiological standards for long-term protection of the public, established by the governmental authorities. The pathways analysis and scenarios give a systematic way to evaluate the potential routes by which people could be exposed to radiation. The scenarios depend on the environment and system characteristics, and on events and processes which could either initiate release of radionuclides from waste or influence their fate and transport to humans and the environment. The choice of appropriate scenarios, pathways and associated conceptual models is very important and strongly influences subsequent analysis of the waste disposal system. This kind of analysis can be used in order to predict the performance of a specific disposal facility and also to derive the total activity permitted to be disposed by radionuclide.

In order to guarantee the radioactive waste will be disposed in a safe way, an analytical model [4] using decay chain series was used by the authors in the geosphere, coupled with the biosphere models and together with a risk assessment. A computer code was developed using the symbolic computation software MATHEMATICA [5]. The results for the groundwater concentration using the analytical model were compared with the numerical simulation by using the MATHEMATICA built-in function NDSolve. The numerical simulation results showed a good agreement with the analytical solution technique here employed.

3.1. Geosphere models and exposure assumptions

The performance assessment of the disposal facility in this study is carried out using the leaching scenario or off-site scenario (considered a normal evolution scenario). This corresponds to the use of contaminated water in the biosphere compartment at the interface with the aquifer, after migration of the radionuclides through the unsaturated and saturated zones. In the interface between the geosphere and the biosphere there is a well intercepting the radioactive plume, at an off-site location where the concentration is the highest (e.g. at the downstream waste site boundary). Accordingly, the biosphere can be composed of a small farm system where the well water is used for drinking and in the production of vegetables, milk, meat and fish. Once the water is used to irrigate, the public can also receive a dose from accidental ingestion of contaminated soil, re-suspended dust and inhalation, external exposure and radon inhalation.

Problems of solute transport involving sequential first-order decay reactions occur in soil and groundwater. This paper presents an analytical model for the vertical transport of radionuclides through the landfill, assuming that all leachate from the landfill is homogeneous. It is also assumed that the release rate from the repository into the unsaturated/saturated media for each member is proportional to the amount remaining in the repository. The total amount of each radionuclide in the waste site as a function of time is $M_i(t)$, and can be determined solving a set of ordinary differential equations [6], shown below, with $M_i(0)$ as the initial amount for each chain member per unit cross-sectional area perpendicular to the direction of flow:

$$\frac{dM_1(t)}{dt} = -\gamma_1 M_1(t) - \lambda_1 M_1(t) \tag{1}$$

$$\frac{dM_i(t)}{dt} = -\gamma_i M_i(t) - \lambda_i M_i(t) + \lambda_{i-1} M_{i-1}(t), \quad \text{for } i = 2, \ldots, n \tag{2}$$

with

$$\gamma_i = \frac{\text{Inf}}{H(\theta + \rho K d_i)} \tag{3}$$

where θ is the moisture content of waste ($L^3\,L^{-3}$), ρ the bulk density of waste ($M\,L^{-3}$), γ_i the leaching rate for each radionuclide (T^{-1}), λ_i the radioactive decay rate (T^{-1}), Inf is the rate of water infiltration through the waste ($L\,T^{-1}$), H the height of waste layer (L), Kd_i the waste form distribution coefficient ($M^{-1}L^3$), t the time (T) and the subscript i stands for the ith member of the decay chain. Then, the hazardous material that leaves the waste site and enters the geosphere is given by $\gamma_i M_i(t)$.

A simplified model for the unsaturated zone can be used, supposing steady-state flow and assuming no dispersion/diffusion. The flow is one-dimensional through a homogeneous zone. The travel time of radionuclide to the water table can be expressed as

$$t_{\text{unsat}} = \frac{H_{\text{unsat}}}{\text{Vi}_{\text{unsat}}}$$

where H_{unsat} is the distance to the water table and Vi_{unsat} is given by $\text{Inf}/(\theta_{\text{unsat}} + \rho_{\text{unsat}} Kd_{i\text{unsat}})$ (the subscript unsat refers to the unsaturated medium). So, the concentration in the vadose zone can be found to be

$$F_i(t) = \gamma_i M_i(t) \exp(-\lambda_i t_{\text{unsat}}) \tag{4}$$

The one-dimensional transport equation for the decay chain in the groundwater can be expressed as [4]:

$$R_1 \frac{\partial C_1(z,t)}{\partial t} = D \frac{\partial^2 C_1(z,t)}{\partial z^2} - v \frac{\partial C_1(z,t)}{\partial z} - \lambda_1 R_1 C_1(z,t) \tag{5}$$

$$R_i \frac{\partial C_i(x,t)}{\partial t} = D \frac{\partial^2 C_i(z,t)}{\partial z^2} - v \frac{\partial C_i(z,t)}{\partial z} - \lambda_i R_i C_i(z,t) + \lambda_{i-1} R_{i-1} C_{i-1}(z,t) \tag{6}$$

where C is the solution concentration ($M\,L^{-3}$), v the average pore-velocity (=q/θ), D the dispersion coefficient ($L^2\,T^{-1}$), q the Darcy velocity ($L\,T^{-1}$), z the distance downward (L) and R_i is the retardation factor given by $R_i = 1 + \rho Kd_i/\theta$, with the following boundary and initial conditions:

$$C_i(0,t) = f_i(t), \quad t > 0 \tag{7a}$$

$$\frac{\partial C_i(\infty,t)}{\partial z} = 0, \quad t > 0 \tag{7b}$$

$$C_i(z,0) = 0, \quad z \geq 0 \tag{8}$$

where $f_i(t)$ is given by $F_i(t)/q$. In this study the solution of the partial differential system (5)–(8) was obtained through the use of the Laplace Transform, as detailed by van Genuchten [4].

In the test case it is assumed that the TENORM waste was placed in the landfill in a layer of 10 m thick and overlain by a thin layer of clean soil. A compacted clay liner is underlining the waste layer and the unsaturated layer is coupled with the aquifer.

3.2. Biosphere models and scenarios

In order to model the biosphere, the small farm scenario will be used, that is, the existence of a farm near the site (at the border) using water from a well for: (a) ingestion of well water; (b) irrigation and: (b1) resuspension and inhalation; (b2) external radiation exposure; (b3) consumption of home grown produce; (b4) consumption of contaminated meat; (b5) ingestion of contaminated milk; (b6) accidental ingestion of contaminated soil; (b7) inhalation of radon and decay products from soil; (c) surface water contact, transfer to fish and to man.

The following equations were used to estimate the annual dose ($Sv\,y^{-1}$), according to the scenario described above

$$\text{DCEN}_{\text{water}} = Q_{\text{water}} C_{\text{water}}\, \text{FCD}_{\text{ing}} \times 10^3 \tag{9a}$$

$$\text{DCEN}_{\text{inh}} = A_{\text{soil}} b_r R_{\text{dust}} \%_{\text{occ}} \times 8766\, \text{FCD}_{\text{inh}} \tag{9b}$$

$$\text{DCEN}_{\text{ext}} = A_{\text{soil}} \times 8766\, \text{FCD}_{\text{ext}} \%_{\text{occ}} \tag{9c}$$

$$\text{DCEN}_{\text{veg}} = (Q_{\text{leg}}\, \text{FT}_{\text{leg}} + Q_{\text{veg}}\, \text{FT}_{\text{veg}}) A_{\text{soil}} f_{\text{soil}}\, \text{FCD}_{\text{inh}} f_{\text{red}} \tag{9d}$$

$$\text{DCEN}_{\text{meat}} = Q_{\text{meat}} \{C_{\text{water}} q_{\text{water}} + A_{\text{soil}} q_{\text{soil}} + A_{\text{soil}} q_{\text{pasture}}\, \text{FT}_{\text{grass}}\}\, \text{FT}_{\text{meat}}\, \text{FCD}_{\text{ing}} f_{\text{red}} \tag{9e}$$

$$\text{DCEN}_{\text{milk}} = Q_{\text{milk}} \{C_{\text{water}} q_{\text{water}} + A_{\text{soil}} q_{\text{soil}} + A_{\text{soil}} q_{\text{pasture}}\, \text{FT}_{\text{grass}}\}\, \text{FT}_{\text{milk}}\, \text{FCD}_{\text{ing}} \tag{9f}$$

$$\text{DCEN}_{\text{soil}} = A_{\text{soil}} Q_{\text{soil}}\, \text{FCD}_{\text{ing}} \tag{9g}$$

$$\text{DCEN}_{\text{radon}} = C_{\text{Rn}} \times 8766\, \%_{\text{Rn}} K_1 K_2 f_{\text{eq}} \tag{9h}$$

$$\text{DCEN}_{\text{fish}} = C_{\text{water}} Q_{\text{fish}}\, \text{FT}_{\text{fish}}\, \text{FCD}_{\text{ing}} f_{\text{red}} \tag{9i}$$

where C_{water} is the concentration in the well ($Bq\,cm^{-3}$), Q_{water} the water ingestion rate ($730\,l\,y^{-1}$), FCD_{ing} the ingestion dose conversion factor ($Sv\,Bq^{-1}$). For the inhalation dose, A_{soil} is given by $C_{\text{water}}\,\text{Irrig}/(\rho_{\text{soil}}\,\text{esp}_{\text{soil}})$, where Irrig is the irrigation rate ($20\,cm\,y^{-1}$), ρ_{soil} the soil density ($1.5\,g\,cm^{-3}$), esp_{soil} the thickness of contaminated soil (15 cm) and b_r is the human breathing rate ($1.0\,m^3\,h^{-1}$), R_{dust} the inhalable outdoor dust ($1.92 \times 10^{-4}\,g\,m^{-3}$), $\%_{\text{occ}}$ the percentage of time spent at place

(40 %) and FCD_{inh} is the inhalation dose conversion factor ($Sv\,Bq^{-1}$).

Regarding the external dose, FCD_{ext} is the external dose conversion factor ($(Sv\,h^{-1})\,(Bq\,g^{-1})^{-1}$), and for the ingestion dose Q_{leg} and Q_{veg} are the root and green vegetables consumption (118 and 20 kg y^{-1}, respectively), FT_{leg} and FT_{veg} are the soil to plant concentration factors for root and green vegetable, f_{soil} the interception factor (0.33) and f_{red} is the preparation reduction factor for food (0.5).

For meat ingestion, Q_{meat} is the consumption of cow meat (63 kg y^{-1}), $q_{pasture}$ the daily pasture intake (68 kg d^{-1}), FT_{grass} the transfer coefficient soil/plant for pasture and FT_{meat} is the transfer coefficient for meat (day kg^{-1}). Q_{milk} is the consumption of cow milk (72 l y^{-1}) and FT_{milk} is the transfer coefficient for milk (day l^{-1}). Q_{soil} is the inadvertent consumption of soil (36.5 g y^{-1}) and Q_{fish} is the consumption of freshwater fish (5.4 kg y^{-1}), FT_{fish} is the concentration ratio for freshwater fish ($cm^3\,kg^{-1}$).

Finally, for the radon calculation, C_{Rn} is the external radon concentration ($Bq\,m^{-3}$), K_1 the effective dose equivalent corresponding to an absorbed energy of 1 J (1.1 Sv J^{-1}), K_2 the potential energy (J m^{-3}) for 1 Bq of Rn-222 in equilibrium with its daughters (5.54×10^{-9} J Bq^{-1}) and f_{eq} is the equilibrium factor (0.8). The ingestion, inhalation and external dose conversion factors ($Sv\,Bq^{-1}$) can be taken from the Basic Safety Standards [7] and the FT's (transfer coefficients and concentration factors) can be obtained from Refs. [8,9].

For the cancer risk a factor of 0.05 Sv^{-1} is used for radioactive materials, and adopts an annual risk of 5×10^{-6} for a dose of 0.1 mSv y^{-1}. The risk is obtained multiplying the estimated annual dose by 0.05. To evaluate the risk according to the pathways, we have used

$$\text{risk}_{inh} = 0.05[\text{DCEN}_{inh} + \text{DCEN}_{radon}] \tag{10a}$$

$$\text{risk}_{ing} = 0.05[\text{DCEN}_{water} + \text{DCEN}_{veg} + \text{DCEN}_{meat} + \text{DCEN}_{milk} + \text{DCEN}_{fish} + \text{DCEN}_{soil}] \tag{10b}$$

$$\text{risk}_{ext} = 0.05\,\text{DCEN}_{ext} \tag{10c}$$

4. Results

As an example of the described methodology, the disposal of slag produced by pyrometallurgy is simulated, with typical concentrations and activities of U decay series. The industrial landfill characteristics are: thickness = 10 m, $\rho = 2.0$ g cm^{-3}, Inf = 0.50 m y^{-1}, $\theta = 0.3$, with the following activities concentration: $C_{U238+234} = 80$ Bq g^{-1}; $C_{Th} = 25$ Bq g^{-1} and $C_{Ra} = 23.5$ Bq g^{-1}. Different simulations were performed, one considering the waste placed directly above the aquifer, another with a compacted clay liner (thickness of 1 m) and a sand vadose (thickness of 3 m) between the waste and the saturated zone. The well is located 100 m from the border of the repository. The data for the *Kd* that were used are given in Table 1.

Table 1
Geosphere *Kd* ($cm^3\,g^{-1}$)

Nuclide	Waste	Liner	Vadose	Aquifer
U	50	1500	560	10
Th	3300	5400	3000	500
Ra	100	9000	500	200

A critical factor in the transport of radionuclides by water through the unsaturated and saturated zone is it retardation by the geologic media, characterized by the *Kd*. The higher this coefficient, the greater the hold-up. Since the source leaching rate and the distribution coefficients are two of the most critical parameters affecting the results for the groundwater water-related pathway, site-specific values should be obtained if the estimated doses or risk approach regulatory limits.

The small farm scenario was modelled by assuming that rainfall percolated vertically downward through the disposal landfill, the liner and the vadose zone and then, finally, moved rapidly into the aquifer. Radionuclides transported from the waste repository via subsurface groundwater are intercepted by the well and also discharged directly into the stream. The final assumption for this example was that the water from the well was the only source of water available to the resident farmer, and all the fish consumed comes from a nearby stream.

Fig. 1 shows the effect on the radionuclide concentration when a liner and vadose are considered between the waste and the aquifer. The uranium is responsible for the higher concentration present in the groundwater, but is insensitive as to whether there is a liner and vadose zone or not. The radium is the most sensitive radionuclide from the simulation run considering the presence or not of additional layers between the waste and the saturated zone.

In order to demonstrate the relevance of calculating not only the concentration but also the final dose and the associated risk, Fig. 2 gives the scenario risk per radionuclide. It is clear that,

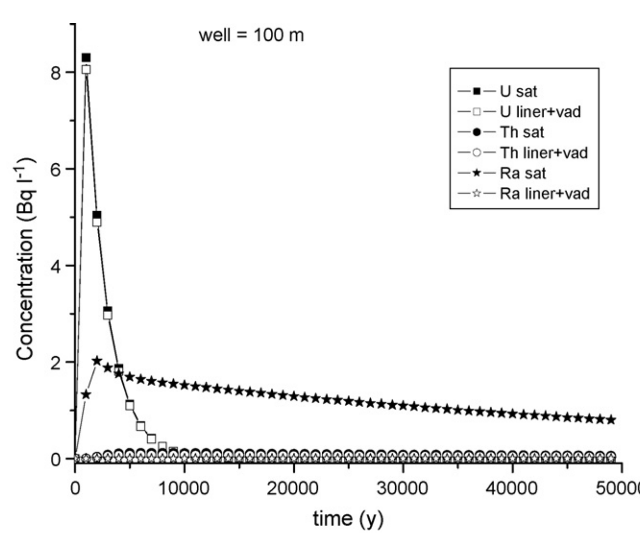

Fig. 1. Influence of liner and vadose zone in the aquifer concentration.

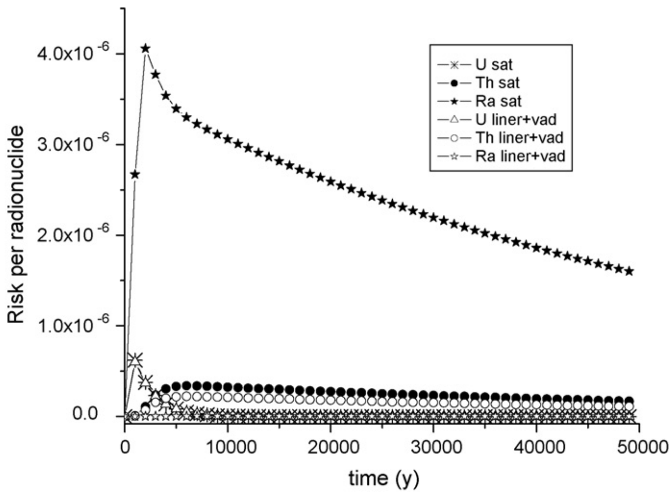

Fig. 2. Time evolution of the risk offered by each radionuclide.

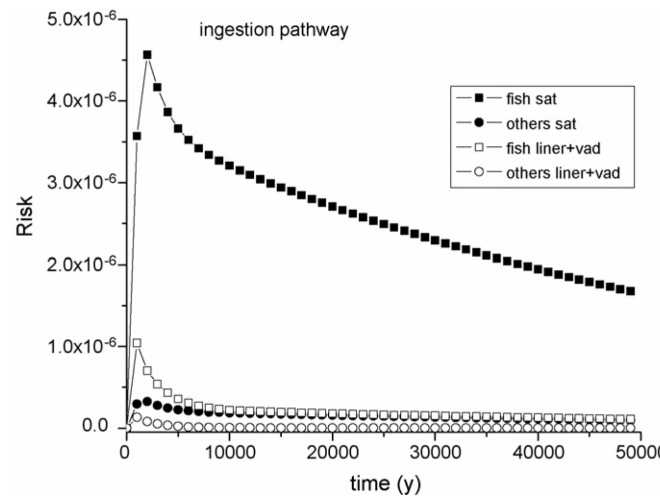

Fig. 4. Time evolution of ingestion pathway risk.

even though the uranium has the highest concentrations, the radium offers the most significant risks, with or without liner and vadose zone.

Fig. 3 illustrates the risk considering the different exposure mechanisms (ingestion, external exposure and inhalation of dust and radon) that can reach human beings. According to these results, ingestion is the most important pathway. The total risk without the liner and vadose zone is close to the waste acceptance criteria adopted (5.0×10^{-6}). This value is reduced by a factor of 5 when the two media between the waste and the groundwater are introduced and accounted for. Again, the figure confirms that ingestion is the most relevant exposure mechanism in this safety assessment.

In order to know which pathway is the most important regarding the ingestion mechanism, Fig. 4 gives the comparison among all the defined pathways, and shows fish ingestion is responsible for the highest dose and risk. This result is of the same order of magnitude as the total ingestion risk. Again, the introduction of additional layers above the groundwater decreases the final dose, illustrating the importance of relevant mitigation strategies in the final safety assessment results.

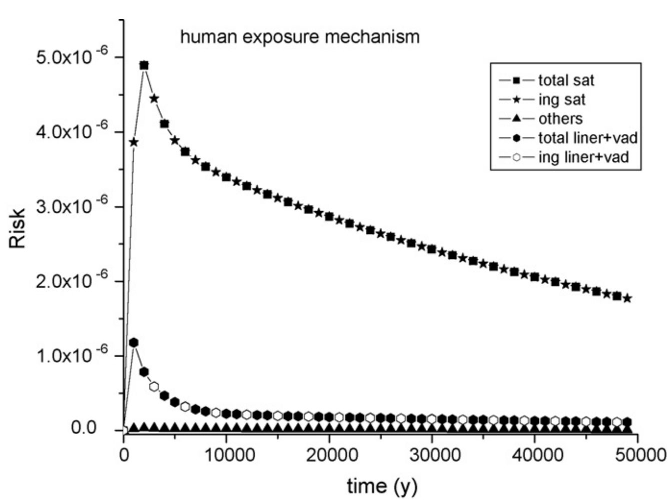

Fig. 3. Time evolution of the risk for each human exposure mechanism.

5. Conclusions

In the present work it was demonstrated that the proposed methodology can be used to judge the ability of the waste landfill design to meet the radiological standards for protection of the public, as established by the national authorities. This kind of evaluation is also useful in examining the effect on performance of various assumptions about confinement capability with time, depending on the barriers used in the repository conception. Even though in many countries the regulatory agency can define the permitted concentration into the groundwater, this study demonstrates that it is very important to define the scenarios and pathways, in order to evaluate the final dose and the associated risks.

The risk assessment performed shows that the radium is responsible for the highest risk that is related to the *Kd* value and the ingestion dose conversion factor. The results for this radionuclide demonstrate that Ra-226 is very sensitive to the presence of barriers, which gives the possibility to reduce the risk by using different layers between the waste and the aquifer.

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