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Preliminary post-closure safety assessment for a borehole-type repository for disused sealed radioactive sources in Brazil

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ABSTRACT

Brazil has a relatively large inventory of disused sealed radioactive sources (DSRSs). Until now, no decision has yet been made about the final destination of this category of radioactive wastes, although a repatriation of a small fraction of these sources comprising mainly neutron and high activity sources was already carried out. Borehole type repositories are one disposal solution considered for DSRSs in Brazil. This paper addresses a preliminary post-closure safety assessment for such a facility, using the borehole disposal concept (BDC) applied to different geological conditions and a range of projected inventories. Results from running the AMBER code considering deterministic and stochastic approaches showed that Am-241 is the main source of potential concern in order to comply with the effective dose constraint of 0.3 mSv/y and allowed the establishment of the relation between the maximum Am-241 inventory and the hydraulic conductivity of the geosphere.

1. Introduction

In many countries worldwide sealed radioactive sources have been in use for many decades in medicine, industry and research for various applications (IAEA, 2005, 2014). Medical facilities represent a large user group for sealed radioactive sources, most notably used for radiotherapy purposes. In industry, sealed radioactive sources are commonly used for radiography applications, quality control measures, well logging, energy supply in remote locations and industrial irradiators, among others. With respect to research, radioactive sources are found in irradiators for conducting radiobiological studies and are used in material science. Other applications of radioactive sources have been ionization smoke detectors and radioactive lightning conductors installed in many countries.

After a useful lifetime of usually 5–15 years the radioactive sources are termed as 'spent' or 'disused' (IAEA, 2005, 2014). Like the varied characteristics of the disused sealed radioactive sources (DSRSs), the possible options for appropriate disposal are diverse (IAEA, 2005). Optionally combined with a preceding decay storage, possible solutions for the disposal of DSRSs are trench or vault type near surface facilities, large cavern facilities at intermediate depth (several tens of meters below surface), shaft/borehole type repositories at depths ranging from ~ 30–300 m and deep boreholes and mined geological repositories of depths greater than 300 m. Both, the activities and the half-lives of the DSRSs as well as their quantities are properties of particular interest when choosing a suitable disposal system. The DSRSs containing radionuclides with higher activities (e.g. Co-60, Sr-90, and Cs-137) and longer half-lives (e.g. Ra-226, Am-241, Pu-238, and Pu-239) require a greater degree of isolation, including but not limited to considerations of human intrusion after an institutional control period of typically 100–300 years, what could be achieved by greater depth and a site and waste specific engineered barrier system (IAEA, 2014).

In the countries of the former Soviet Union (USSR), shallow depth borehole type repositories for the disposal of DSRSs have been in operation for over 40 years (Ojovan et al., 2003). A safety assessment of such a disposal facility was presented in Ojovan et al. (2000). Another example for the relevance of borehole facilities for the disposal of DSRSs are the ongoing works in African countries like South Africa or Tanzania (Salehe and Kim, 2013). These works progressed in conjunction with different projects within the International Atomic Energy Agency's AFRA program (IAEA/AFRA) that started in the beginning of the 1990s. Regarding the management of DSRSs, the first major projects particularly addressed issues associated with the conditioning of spent radium sources in stainless steel capsules of standardized dimensions enabling subsequent handling, transport and storage as well as the disposal of (radium bearing) DSRSs in specially designed borehole disposal facilities (BDF) able to provide long-term safety under a wide range of geological and climatic conditions. The development of a

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portable remote handling device ("mobile hot cell") allowed for the conditioning of DSRSs with higher activities, thus making the borehole disposal concept (BDC) a suitable solution for almost all types of DSRSs (IAEA, 2011).

This paper aims to present the results of a preliminary post-closure safety assessment of the disposal of DSRSs from Brazil in a borehole-type repository using the BDC concept, through a two phase approach. Firstly, a deterministic analysis considering the Brazilian inventory was conducted based on a Generic Safety Assessment (GSA) elaborated for the IAEA's borehole disposal concept for DSRSs. Taking into account the results of the deterministic analysis, a probabilistic sensitivity analysis was developed to identify the response in terms of total dose as a function of the Am-241 inventory and the hydraulic conductivity of the geosphere. This analysis was intended to estimate the limits for the applicability of the BDC concept to the Brazilian situation.

2. The BDC and the GSA

The main safety features of the considered BDC are given below, being consistent with the design described in IAEA (2011):

- Fully welded 3 mm thick 316L stainless steel capsules accommodating the DSRSs. The design provides for capsules with fixed wall thickness and length and two different diameters to encase sources of varying physical sizes.
- Thick walled (6 mm) and 250 mm long, fully welded 316L stainless steel disposal containers. In each disposal container one capsule containing the DSRSs is placed surrounded by a cement buffer, also referred to as containment barrier.
- A narrow diameter (minimum 260 mm at maximum depth) borehole in which the disposal containers are emplaced separated by a concrete backfill in 1 m intervals (Fig. 1). The top of the disposal zone is located at least 30 m below ground/the local erosion base accounting for human intrusion and changes of the geomorphology, respectively. The disposal zone reaches depths of more than 100 m and ends with a 0.5 m long concrete plug in the bottom. An anti-intrusion plate is installed at the top of the disposal zone and the remaining part above is backfilled with concrete. The chosen host rocks will preserve favorable geological conditions and thereby the integrity of the disposal containers for the required period of time (up to tens of thousands of years).

Although the BDC considered for the disposal of DSRSs includes several standardized components and features, adaptions to a specific inventory and/or disposal site characteristics are possible bearing in mind the limitations imposed by the safety assessment. Possible alterations are for example capsules and disposal containers manufactured in diverging lengths, disposal containers accommodating more than one capsule, a reduced spacing between the packages and therefore a relative greater quantity of disposal packages per borehole (IAEA, 2011) and an extended depth of the disposal zone up to several hundred meters (IAEA, 2017). When determining the depth and length of the disposal zone, at least the minimum depth of the water table allowing for seasonal and longer term variations, the depth of the local erosion base and the depths of suitable host formations, the groundwater flow regime and the geochemical conditions have to be taken into account (IAEA, 2011). In this context, it is noteworthy that the disposal zone must not overlap with the interface of the unsaturated and the saturated zone but must be located completely in either the one or the other zone in order not to impair the corrosion resistance of the disposal system and to facilitate the modeling of the near field evolution (IAEA, 2009).

A Generic Safety Assessment (GSA) was developed considering different geosphere configurations and release mechanisms: disposal zone (unsaturated and saturated), release mechanism (gaseous, liquid and solid), flow conditions (low, medium and high flow in porous system and high flow in fracture system) (Little et al., 2004; IAEA, 2017). Besides the design scenario, four different defect scenarios were also considered, including a defect weld closure of one waste container (D1), a defect weld closure of one waste capsule (D2), degraded/incomplete disposal/disturbed zone cement grout (D3) and one waste capsule having a defect weld closure within a waste container with defect weld closure (D4). The GSA model has been implemented in a version of the AMBER software tool (version 4.5), a commercial software tool developed by Quintessa Ltd. AMBER uses a compartment model approach to represent the migration and fate of contaminants in the environment.

IAEA (2017) presents the results for the calculation cases in terms of the activity limits for each radionuclide for which the total dose does not exceed the dose constraint of 0.3 mSv/y. The results have shown that the most restrictive activity limits were obtained for the saturated disposal zone with high flow rate in a porous system, with Pu-238, Pu-239 and Am-241 being the more limiting radionuclides. With respect to the design scenario for disposal in saturated, porous media with high

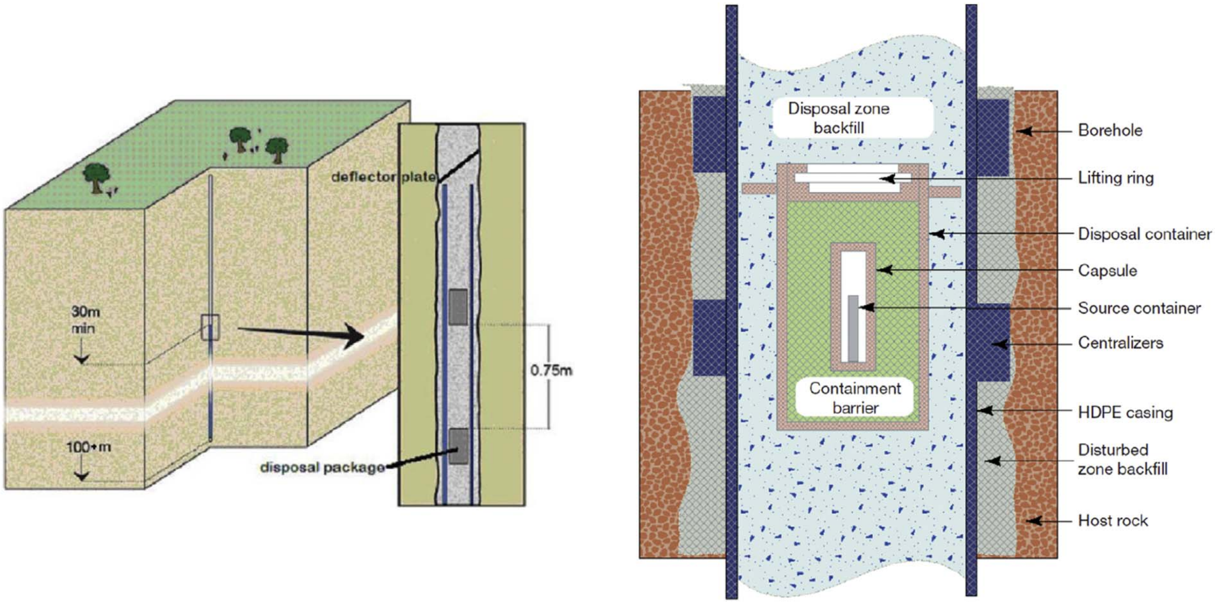

Fig. 1. Scheme of a borehole disposal facility (IAEA, 2011) (left) and illustrative section through the borehole (right) (IAEA, 2009). (Reproductions with permission by IAEA).

flow rate, the total activity limit for Am-241 calculated in the GSA for liquid release is 5.0E+12 Bq. For medium flow rate in similar conditions, the activity limit for Am-241 is 5.0E+14 Bq and for the low flow rate case, no activity limit is given in the GSA (higher than 1.0E+18 Bq). Accounting for the combination of the consequences of the design and defect scenarios, the activity limits for Pu-238 and Pu-239 are 2.0E+12 and 5.0E+11 Bq, respectively. Regarding Am-241, the activity limit calculated in the GSA is 3.0E+12 Bq for the disposal in saturated, porous media with high flow rate and in the order of 1.0E+13 Bq for all other cases. It is important to note that these results did not take in consideration any possible thermal or radiolysis effects.

3. Materials and methods

3.1. Inventory

A total of 29 radionuclides have been identified in intermediate storage facilities for disused radioactive sources in Brazil (Aguiar et al., 2015). The majority of the sources are of Am-241 (91.2%) while in terms of activity, Co-60 sources dominate (97.2%). As in IAEA (2017), the inventory was reduced by screening out any radionuclides that, due to their half-life, maximum activity, or radiotoxicity, will not result in significant post-closure impacts and therefore do not have to be explicitly considered in the safety assessment. The number of sources and the initial inventory of the 19 radionuclides remaining after the screening process are presented in Table 1.

It is important to consider that no attempt was made in this study to adjust the dimensions of the borehole components to allow physically fitting of all the Brazilian inventory. The total quantity and volume of the sources to be disposed of are not predictable at present, due to the unknown accurate dimensions of the most of the sources, either due to the not yet clear country DSRS repatriation police. Therefore, it is assumed that the quantity and dimensions of the inventory sources are compatible with the reference borehole configuration. However, this is a crucial consideration that must be throughly verified in further developments of the work.

3.2. Modeling process

Considering the current Brazilian inventory and the GSA results, the only radionuclide of concern would be Am-241 in case of a saturated disposal zone in porous media with high flow rate, remembering that this statement did not take in consideration any possible thermal or radiolysis effects due to the Cat. 1 & 2 Co-60, Cs-137 and Am-241 sources. Therefore, this analysis focuses on investigating the radiological consequences of disposing of the Brazilian DSRSs in a saturated sedimentary formation, including the design and the four defect scenarios proposed in IAEA (2017). For this study, the same model developed and implemented for the GSA served as the basis for the Brazilian safety assessment calculations. Also following the GSA modeling process, the simulation period was set as to allow the time of peak impacts to be evaluated.

Table 1
Brazilian inventory of disused radioactive sources (at December 2014).

Radionuclide	Quantity (-)	Initial activity (Bq)	Half-life (y)
Am-241[a]	169,506	1.75E+13	4.32E+02
Ba-133	7	1.02E+07	1.07E+01
Cf-252	15	2.76E+08	2.64E+00
Cm-244	7	8.34E+09	1.81E+01
Co-60	2093	8.58E+14	5.27E+00
Cs-137	3098	9.59E+13	3.00E+01
Fe-55	57	2.73E+09	2.70E+00
H-3	118	7.93E+11	1.24E+01
K-40	1	6.29E+04	1.28E+09
Kr-85	327	1.94E+12	1.07E+01
Ni-63	121	7.78E+10	9.60E+01
Pm-147	227	5.57E+10	2.62E+00
Pu-238	4	3.11E+09	2.41E+04
Ra-226[b]	8067	5.83E+11	1.60E+03
Sr-90	271	1.46E+11	2.91E+01
Th-232	3	1.39E+05	1.40E+10
Ti-44	1	2.17E+07	4.73E+01
Tl-204	63	5.14E+08	3.78E+00
U-238	1	8.51E+03	4.47E+09

[a] Comprises Am-241 and AmBe-241 sources and Am-241 tapes from lightning rods and smoke detectors.

[b] Comprises Ra-226 and Ra-226-Be sources and Ra-226 tapes from lightning rods and smoke detectors.

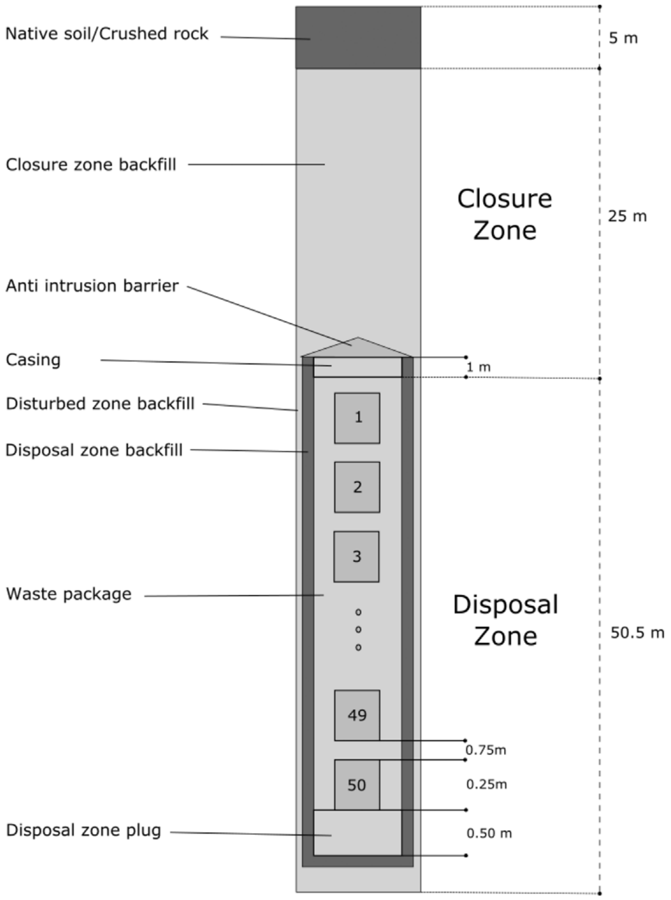

Fig. 2. Near field configuration for liquid release in saturated disposal zone (not to scale).

3.3. Modeling parameters

Geometry information of the near field for all simulations are presented in Fig. 2. The doses are calculated based on a scenario where water from an abstraction well located 100 m downstream from the borehole is used for human and animal consumption and irrigation. As in the GSA model, dose coefficients include contributions of short-lived (half-life less than 25 days) daughters not explicitly listed, assuming secular equilibrium at time of intake or exposure. Internal and external dose coefficient were obtained mainly from the Brazilian regulations (CNEN, 2011), unless not available. In these cases, as well as for the element-specific data, values were obtained from IAEA (2017). When data were not available in this report, literature data were used. A detailed description of all parameters and the respective values can be found in Aguiar et al. (2015).

For the probabilistic sensitivity analysis, two parameters were considered as sampled variables: host-rock hydraulic conductivity and Am-241 activity. This decision was based on the results of the

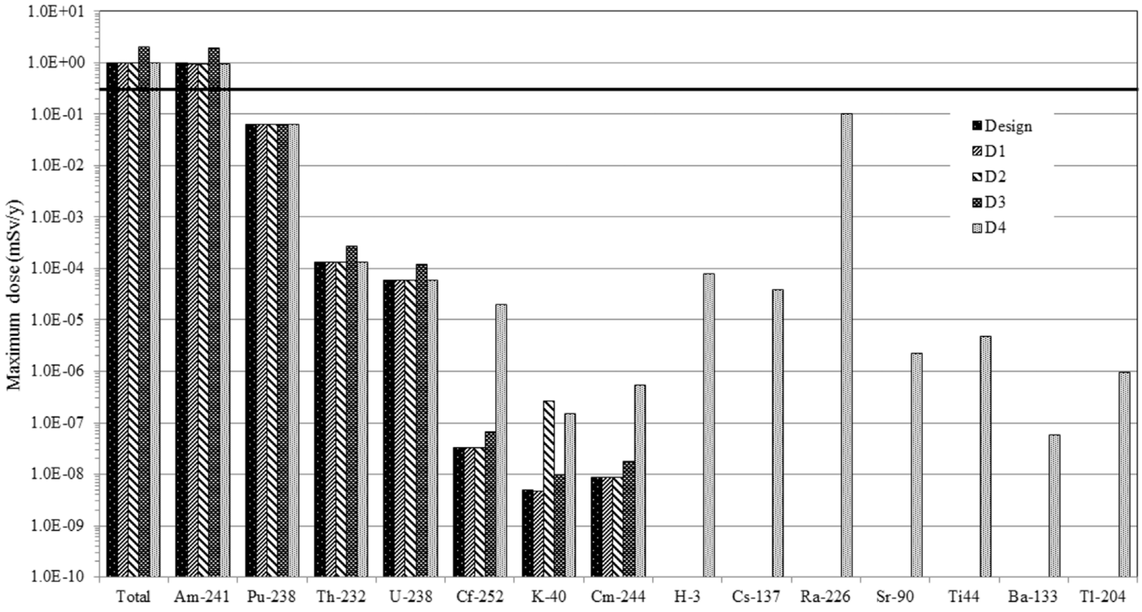

Fig. 3. Maximum doses (total and for each chain) for the calculated cases involving high flow rates.

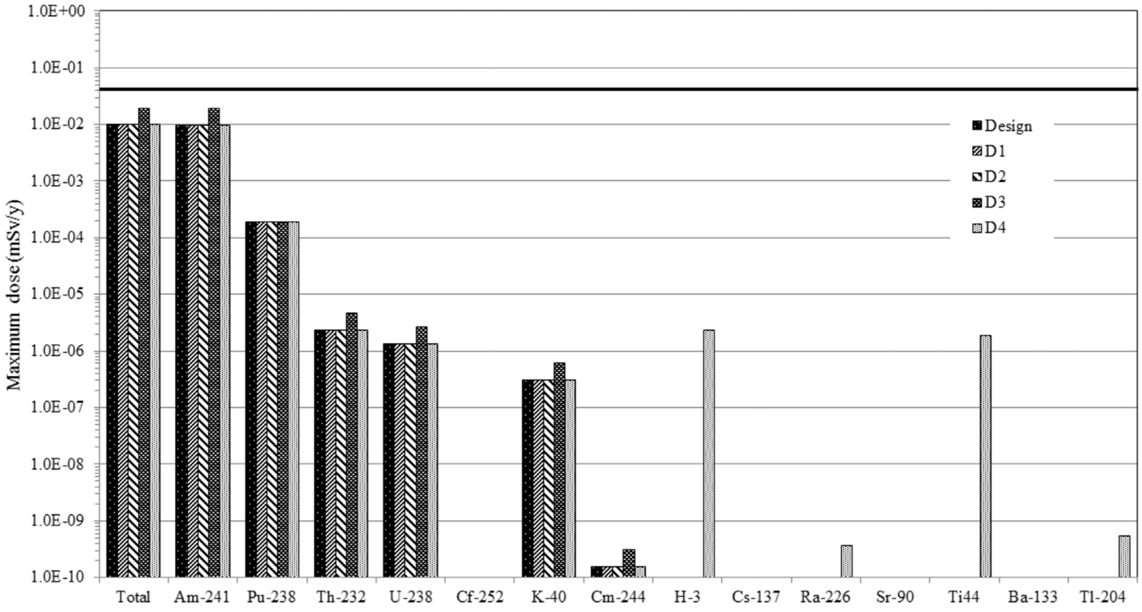

Fig. 4. Maximum doses (total and for each chain) for the calculated cases involving medium flow rates.

deterministic analysis as will be discussed later. In order to simplify the analysis, the time of failure of the engineered barriers, which are flow rate-dependent, were kept constant as for the high flow scenario. All other variables and parameters were maintained as discussed for the deterministic analysis.

This simulation has its focus on porous formations as possible host-rocks for the repository. A range of hydraulic conductivity values was selected based on data presented in Davis (1969) and Domenico and Schwartz (1997). Based on these references, the maximum hydraulic conductivity value of shale rocks was adopted as the minimum value for the sensitivity analysis: 6.3E-02 m/y (2.0E-09 m/s). This value is compatible with GSA parameters assumed in IAEA (2017) and cover their lowest flow rate, for allowing the comparison of the results. The maximum value of hydraulic conductivity was set as 1.0E+03 m/y (3.2E-05 m/s) based on the GSA analysis for the high flow rate case. This assumption was conservative since the higher values of hydraulic conductivity found in bibliography was lower than the value assumed in GSA.

The Am-241 activity range was chosen considering the current total activity of 1.75E+13 Bq. An estimation about the maximum activity was made considering the amount of sources in use in Brazil (Aguiar et al., 2015). The maximum expected activity of Am-241, including the current inventory of disused sources, was calculated as 6.5E+13 Bq. The minimum value for the Am-241 activity was set as one order of magnitude lower than the current activity for taking account the possibility of a future repatriation of some of the Am-241 sources (1.75E+12 Bq).

For the probabilistic sampling of both hydraulic conductivities and Am-241 inventory, a uniform probability density function (PDF) was assumed, meaning that equal probability was assigned for each sample in the value range of the stochastic variables. Latin Hypercube Sampling (LHS) was used as the sampling method to assure that the whole interval of concern for the parameter values were considered (Helton and Davis, 2003). The number of samples was set to 1000 for

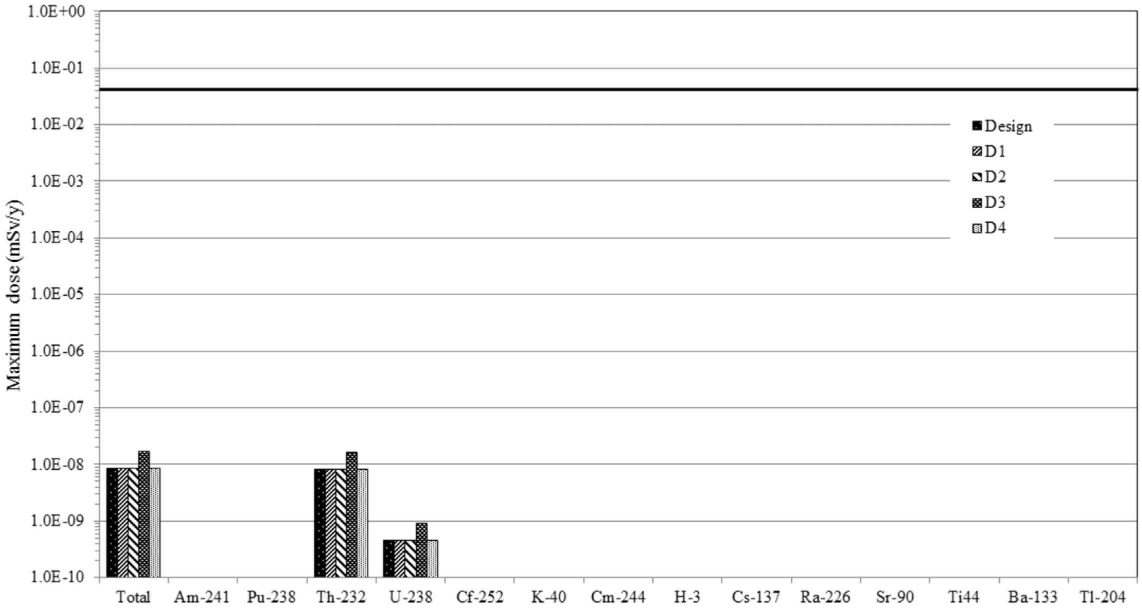

Fig. 5. Maximum doses (total and for each chain) for the calculated cases involving low flow rates.

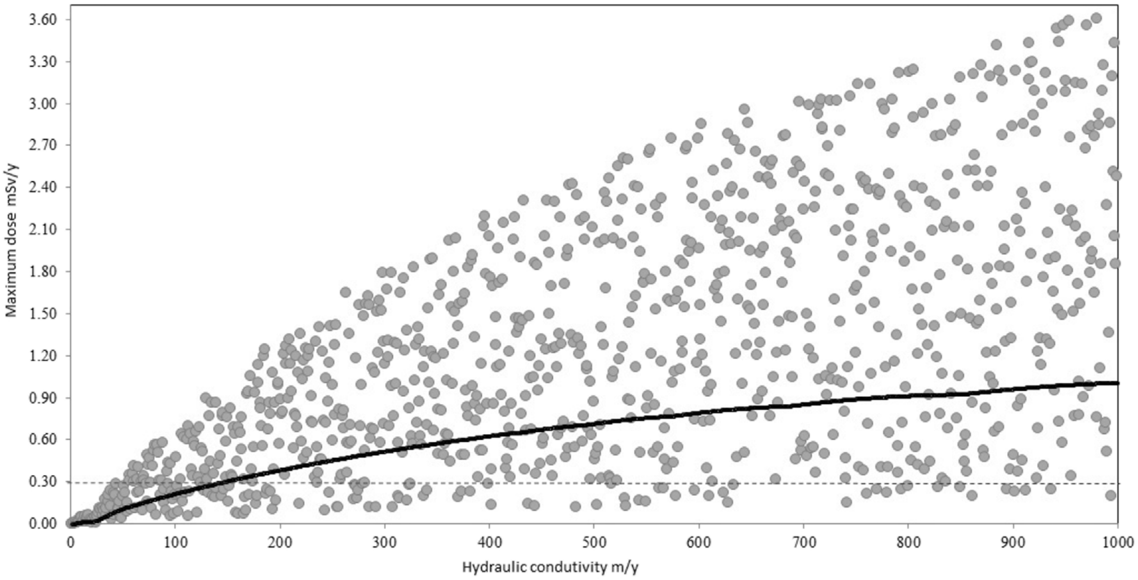

Fig. 6. Maximum total doses versus hydraulic conductivity for each sampled Am-241 activity (grey dots). The dashed line marks the dose constraint and black line represents the results for the deterministic Am-241 activity.

the simulation.

As will be discussed later, the deterministic simulations showed that there is no significant difference between the design and defect scenarios for a given flow rate system. Based on this result, the sensitivity analysis was carried out only considering the design scenario.

4. Results and analysis

4.1. Deterministic analysis

The maximum effective doses for the design and the four defect scenarios for saturated zone disposal in porous media are presented in Figs. 3–5 for high, medium and low flow rates, respectively, for both the total and for the main radionuclide chains. The dose constraint is indicated in these figures as a horizontal line. For all scenarios, design and defects alike, the results have shown that the only case for which the doses exceed the value of 0.3 mSv/y is for high flow rates in geosphere. For medium and low flow rates, the estimated doses are a factor of two and nine orders of magnitude smaller than the constraint, respectively. The time of the peak doses were 7.4E5, 2E6 and > 1E9 years for high, medium and low flow rates, respectively.

For most calculated cases, the Am-241 chain is the dominant contributor for the total dose (Np-237, Pa-233, U-233 and Th-229), followed by Pu-238 (U-234, Th-230, Ra-226, Pb-210 and Po-210). The short-lived daughters contribution to total dose are considered by assuming secular equilibrium. Note that the results obtained for Am-241, considering the Brazilian inventory of 1.75E + 13 Bq, are consistent with the total activity limit (5E + 12 Bq) calculated in the GSA report for the design scenario in high flow rate in porous saturated media. The activity of the Brazilian Am-241 sources is lower than the total activity limit predicted in the GSA for medium flow rate in similar geological conditions. For all other radionuclides of the Brazilian inventory, the activity does not exceed the activity limits calculated in the GSA.

Consistent with the GSA results, it can be observed that there is no

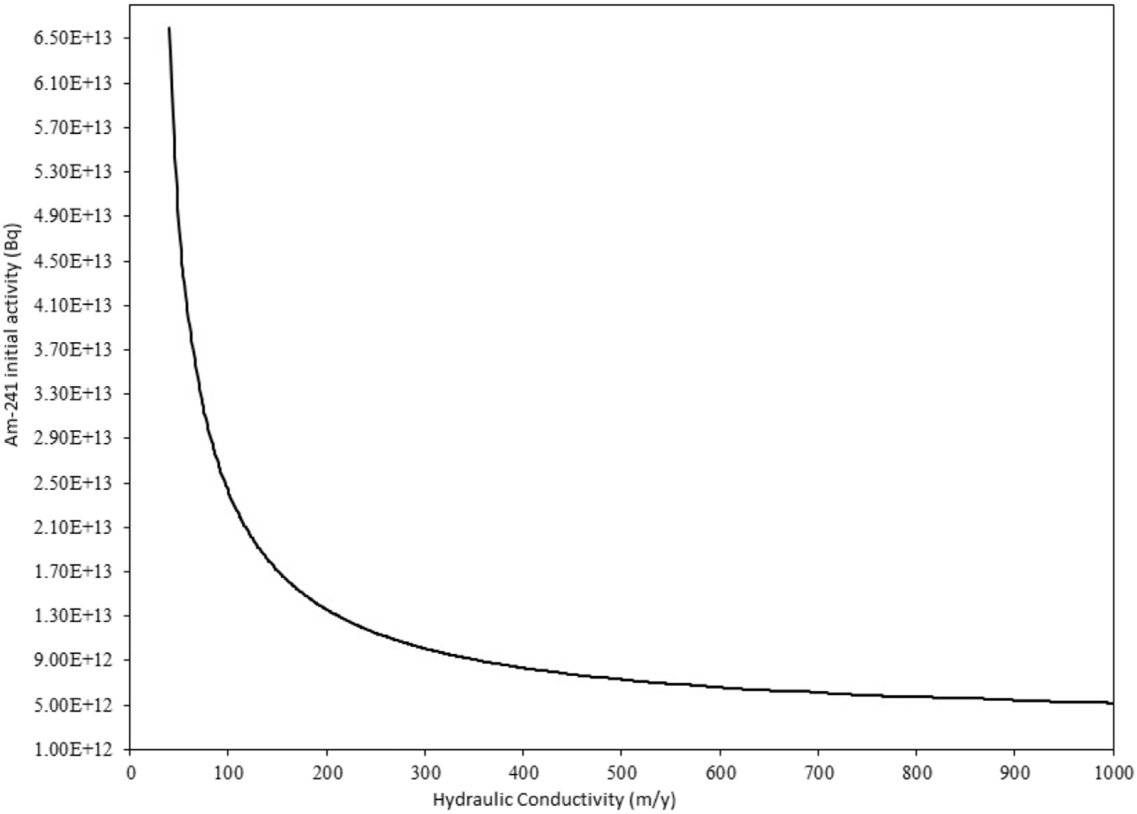

Fig. 7. Maximum Am-241 activities versus hydraulic conductivity sampled values for the sensitivity analysis. Any combination of the parameter values below or on the curve indicate compliance with the dose constraint of 0.3 mSv/y.

significant variation between the doses estimated for the design and defect scenarios cases for a given flow rate in the geosphere. Accordingly to GSA, these results emphasizes the multiple barrier nature of the near field that ensures defects in the performance of one barrier do not compromise the performance of the near field. The only exception is for the defect scenario D4, involving a faulty waste capsule being within a faulty disposal container, for which other radionuclides with lower half-life (for instance, Ra-226, H-3 and Cs-137) play a more relevant role (mainly for high flow rate cases), although still complying to the dose limit criteria. This result is also compatible with those stated in GSA document.

4.2. Probabilistic sensitivity analysis

The probabilistic analysis showed a large model sensitivity for the range of the analyzed parameters. While, as expected, the maximum dose increases linearly with the activity of Am-241, the hydraulic conductivity is related with the maximum dose by a exponential relation with a decimal exponent. Fig. 6 shows the maximum dose for all radionuclides represented as function of the hydraulic conductivity variation. Each grey dot indicates the maximum total doses calculated for a pair of sampled values of Am-241 activity and hydraulic conductivity. In this figure, the dashed line marks the dose constraint (0.3 mSv/y). The black line represents the model results when the Am-241 activity is kept constant as in the deterministic analysis (1.75E + 13 Bq).

Fig. 6 allows the model sensitivity for the sampled combinations of hydraulic conductivity and Am-241 activity values to be analyzed. From the intersection between the black and dashed line it is possible to estimate a maximum value of approx. 140 m/y for the hydraulic conductivity in order to comply with dose constraint considering the current Am-241 inventory. Fig. 7 shows the region below curve that represents combinations of Am-241 activity and hydraulic conductivity that result in a dose smaller than 0.3 mSv/y. The model results allowed to establish the relationship between the possible maximum Am-241 inventory and the evaluated range of hydraulic conductivity in order to comply with the dose constraint (Fig. 7). The curve in this figure shows the limit for the possible combinations of the parameter values for which the model results were below or at the dose constraint. It is observed that values of hydraulic conductivities below 50 m/y will result in compliance with the dose constraint for the considered Am-241 activity range. The same result is obtained for Am-241 activity below 4.5E + 12 Bq for the whole evaluated range of hydraulic conductivity values (Fig. 7). It is important to keep in mind that these results are valid for the assumed failure times for the engineered barriers.

The validity of the results are linked to the specified near-field geometry, transport and consumption scenario configurations implemented in the AMBER code for the current assessment. The main limitation is related to the performance of the engineered barriers, specifically with respect to the time of failure of each barrier. In this analysis, the dependence of these parameters with the groundwater flow rate was not considered. In order to assure the designed performance, the site specific geochemistry conditions, such as high pH and low levels of chlorine and sulphate, must be investigated (IAEA, 2011, 2017). In case of other hydrogeochemical conditions, a specific evaluation of the failure times for the barriers must be carried out. Another limitation is related to restraining the variations in the geosphere flow rate only to the hydraulic conductivity. Variations of the porosity in different host-rocks are usually more limited than hydraulic conductivity but can also contribute to obtain different flow rates.

5. Conclusions

Preliminary results from the deterministic simulations showed that Am-241 sources are of the most concern among the Brazilian DSRS inventory, followed by Pu-238 sources. For all the hydrogeological scenarios simulated, the only case in which the total dose constraint of

0.3 mSv/y is exceeded was associated with high flow rate conditions. By developing a stochastic sensitivity analysis considering two of the expected most relevant variables, namely, hydraulic conductivity and Am-241 activity, it was possible to establish the relation between these variables as limits for complying with the total dose constraint. For the current Am-241 inventory, the maximum hydraulic conductivity is estimated to be around 140 m/y in order to comply with the total dose constraint. In order to be able to dispose of the maximum projected Am-241 inventory, the simulations indicated that the hydraulic conductivity must be below 50 m/y.

For the development of this work, some important assumptions were made: thermal or radiolysis effects were not considered; the assumed BDC design could physically fit the current Brazilian inventory in a single borehole; for the probabilistic calculations, the failure time for the engineered barriers was kept constant as estimated for the high flow scenario. Although the results of this study are preliminary and limited to the scenario configuration and parameter values adopted, they can be used to support future site selection activities.

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