



# Development of sustainable extraction method for long-lived radioisotopes, $^{133}\text{Ba}$ and $^{134}\text{Cs}$ using a potential bio-sorbent

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## Abstract

Common agro-waste, potato peels were used to study the efficacy of adsorption of long-lived radioisotopes,  $^{133}\text{Ba}$  ( $T_{1/2} = 10.54$  a) and  $^{134}\text{Cs}$  ( $T_{1/2} = 2.06$  a) as a precursor of fission products  $^{140}\text{Ba}$  ( $T_{1/2} = 12.75$  d) and  $^{137}\text{Cs}$  ( $T_{1/2} = 30.07$  a). At optimized condition of pH = 3, high  $K_d$  value  $> 3 \times 10^4 \text{ cm}^3 \text{ g}^{-1}$  was observed for Ba, when 60 mg dried potato peel was used as sorbent-material for adsorption from a binary aqueous solution containing  $^{133}\text{Ba}$  and  $^{134}\text{Cs}$ . Cs showed low  $K_d$  values across the entire pH range. The bio-sorbent could effectively uptake ~99% of  $^{133}\text{Ba}$  and ~48% of  $^{134}\text{Cs}$  at the best condition.

**Keywords** Potato peel · Bio-sorption · agrowaste ·  $^{134}\text{Cs}$  ·  $^{133}\text{Ba}$  ·  $\gamma$ -spectrometry

## Introduction

Environmental protection from hazardous materials is always a concern and challenge. Heavy metal pollution remains one of the most explored fields of research since aquatic systems get easily contaminated with industrial-domestic run-offs or effluents thereby distressing the floral-faunal communities. Conventional techniques like ion exchange, reverse osmosis, membrane filtration, etc., have broad application in this field [1, 2]. Besides, environmentally benign and eco-friendly methods like phytoremediation [3–5], use of natural bio-sorbents, bio-degradable materials [6–10], etc., are also providing efficient, safe, economical way of bio-remediation.

Similar to heavy metals, radioactive waste materials pose a serious threat to the environment and its components if not properly stored or managed. Many industrial, medical, research and nuclear sectors regularly use radioactive materials for various purposes. Even at trace concentrations, radioisotopes may exhibit chemical and radiological toxicity to living organisms [11]. In this regards, radioactive waste management is both demanding and challenging.

Radio-caesium is one of the major long-lived elements in liquid radioactive wastes (LRW). Apart from generated wastes, nuclear fall-outs, activities or nuclear accidents contribute significantly towards radioisotopic contamination of the surrounding, which is also definitely a cause for concern [12, 13]. Plenty of research and modeling related to extent of radioactive contamination has been reported following nuclear accidents like Chernobyl, Fukushima-Daichii, etc. [14–19]. In most of the reports, emphasis has been put on scrutinizing the contamination, migration and residence properties of labile caesium ( $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ), which is easily transportable [14, 20–23]. Cs being labile and soluble gets easily transferred upward along the food chain, eventually entering the human system. Hence, several biological, chemical and physical methods have been undertaken to study adsorption or extraction behavior of such long-lived isotopes [24, 25].

Long-lived radionuclides like  $^{133}\text{Ba}$  ( $T_{1/2} = 10.54$  a),  $^{134}\text{Cs}$  ( $T_{1/2} = 2.06$  a) and fission products like  $^{137}\text{Cs}$  ( $T_{1/2} = 30$  a) and  $^{140}\text{Ba}$  ( $T_{1/2} = 12.75$  d), when released in the environment, may lead to radioactive contamination in the surrounding. In fact, both  $^{137}\text{Cs}$  and  $^{140}\text{Ba}$  have high fission yield (Table 1). It is always imperative to develop efficient techniques towards separation or absorption of such radioisotopes. Hence, studies on extraction behavior of caesium and barium radioisotopes are of interest and importance.

In fact, cheaper and accessible substitutes like chitosan, alginates, agricultural wastes, wood wastes, clay materials, etc., are becoming useful sorbent-materials. Calcium

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**Table 1** Fission yields of Cs and Ba radioisotopes [26]

Nuclide	Fission Product	Thermal fission yield (% per fission)	Fast fission yield (% per fission)
$^{232}\text{Th}$	Cs-134	–	$0.00000015 \pm 0.000000041$
	Cs-137	–	$6.3 \pm 0.3$
	Ba-140	–	$7.71 \pm 0.25$
$^{233}\text{U}$	Cs-134	$0.00114 \pm 0.00029$	$0.00158 \pm 0.00041$
	Cs-137	$6.2 \pm 0.22$	$6.5 \pm 0.31$
	Ba-140	$6.43 \pm 0.26$	$6.17 \pm 0.2$
$^{235}\text{U}$	Cs-134	$0.0000121 \pm 0.0000032$	$0.0000279 \pm 0.0000073$
	Cs-137	$6.221 \pm 0.069$	$5.889 \pm 0.096$
	Ba-140	$6.314 \pm 0.095$	$5.959 \pm 0.048$
$^{238}\text{U}$	Cs-134	–	$0.000000113 \pm 0.00000003$
	Cs-137	–	$6.02 \pm 0.5$
	Ba-140	–	$5.972 \pm 0.084$

alginate beads had been studied for uptake of  $^{133}\text{Ba}$  from a binary mixture of  $^{133}\text{Ba}$  and  $^{134}\text{Cs}$  [27]. Radioactive Ba and Cs have been removed by several bio-adsorbents efficiently including mosses, seeds, agricultural wastes, plant materials, bio-materials (beads, chitosan), etc. [28–34].

The aim of present study is primarily to investigate the efficacy of a low-cost, easily available potato-peel (*Solanum tuberosum*) wastes for adsorption of the most labile fission products  $^{137}\text{Cs}$ . However, the fission yield of the adjacent  $^{140}\text{Ba}$  is equal to that of  $^{137}\text{Cs}$ . Therefore, adsorption probability of  $^{140}\text{Ba}$  on potato peel was also considered. We have used binary mixture of  $^{133}\text{Ba}$  and  $^{134}\text{Cs}$ , which would act as precursor of  $^{140}\text{Ba}$  and  $^{137}\text{Cs}$  respectively. In this experiment,  $^{137}\text{Cs}$  has been purposefully avoided as it is in secular equilibrium with  $^{137}\text{Ba}$  ( $T_{1/2} = 2.52$  min), which shares the same  $\gamma$  line with  $^{137}\text{Cs}$  at 661.6 keV and would confuse the adsorption pattern between Ba and Cs radionuclides. The experimental parameters like pH, time of contact, amount of adsorbent, etc., affecting the adsorption behavior have been optimized.

## Materials and methods

### Chemicals and reagents

All the chemicals used for the present study were of analytical grade. De-ionized water (18.2 M $\Omega$  cm) was obtained from ThermoScientific smart2 Pure 6 UV/UF water purification system. Emparta concentrated  $\text{HNO}_3$ , procured from Merck, India was used for the experiment.

Potatoes were purchased from local market and washed thoroughly. Afterwards the peels were separated from potato and were again thoroughly flushed under running tap water

for removal of unwanted dirt. Next, these were rinsed in de-ionized water and thereafter soaked in sufficient volume of 0.1 M  $\text{HNO}_3$  for surface protonation. Cleansed peels were left overnight, completely air-dried, also oven-dried and finally pulverized into fine powder, sieved (mesh no. 100) and used as the bio-sorbent for  $^{133}\text{Ba}$  and  $^{134}\text{Cs}$  adsorption.

$^{133}\text{Ba}$  and  $^{134}\text{Cs}$  radionuclides were procured from Board of Radiation and Isotope Technology (BRIT), Mumbai. A binary stock solution of 5 mL was prepared by mixing  $^{133}\text{Ba}$  and  $^{134}\text{Cs}$ . From this stock, every time 100  $\mu\text{L}$  of activity was taken for each batch of extraction process.

### Instruments

p-type, coaxial well-type high-purity germanium (HPGe) detector (make CANBERRA) having 30% relative efficiency and 2.7 keV resolution at 1332 keV energy was used for all the radiochemical measurements. The detector is placed inside 5 cm thick lead cylindrical rings with 0.2 cm steel shield inside. The associated electronics, DSA-1000 was procured from CANBERRA. Energy calibration of the system was performed using point sources of  $^{152}\text{Eu}$  and  $^{60}\text{Co}$ . Efficiency calibration was done inside the well with a liquid  $^{152}\text{Eu}$  source of known strength to maintain the same geometry of the samples. Gamma-spectra obtained were analyzed using Genie 2000 software. For the batch experiment, 1.5 mL of sample was taken in Eppendorf and counted inside the well, and stripped off the respective backgrounds.  $^{133}\text{Ba}$  was radiometrically monitored from its  $\gamma$ -peak at 81 keV, whereas 604.7 keV  $\gamma$  peaks were considered for  $^{134}\text{Cs}$ .

Functional groups present in the bio-sorbent (PP) was determined by FTIR spectroscopy. IR spectra of dried potato peels in KBr were recorded using a Perkin-Elmer FTIR RX1 spectrometer (4500–500  $\text{cm}^{-1}$ ).

## Radiochemical extraction

A binary stock solution of 5 mL was prepared by mixing  $^{133}\text{Ba}$  and  $^{134}\text{Cs}$ . From this stock, every time 100  $\mu\text{L}$  of activity was taken for each batch of extraction process. All measurements were done inside the well of the HPGe detector. This allowed us to work with minimum amount of radioactivity. The activity of  $^{134}\text{Cs}$  and  $^{133}\text{Ba}$  in each 100  $\mu\text{L}$  solution were 64 and 200 Bq, respectively.

Adsorptions of  $^{133}\text{Ba}$  and  $^{134}\text{Cs}$  were studied in batch mode by powdered potato peel (PP). Solid liquid extraction (SLX) was performed with 3.9 mL  $\text{HNO}_3$  solution of different concentrations, (0.0001, 0.001, 0.01, 0.1 and 1 M) as aqueous phase and 20 mg PP as solid phase. 0.1 mL binary stock solution containing  $^{133}\text{Ba}$  and  $^{134}\text{Cs}$  was added to experimental samples, mechanically shaken for 10 min followed by 10 min centrifugation at 5000 rpm. After phase separation, 1.5 mL supernatant was taken in Eppendorf for gamma-spectroscopic study.

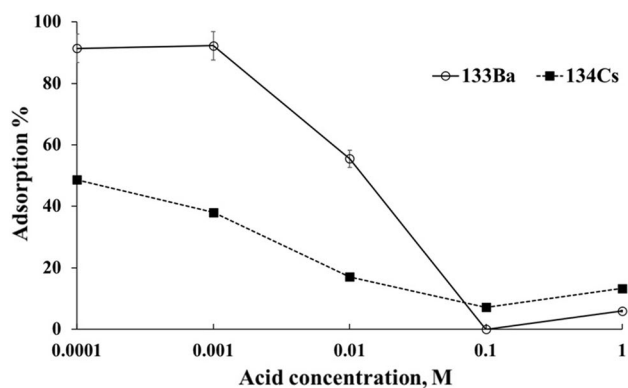
Shaking time (5, 10, 20, 30, 40, 60 min) was varied keeping the amount of activity (0.1 mL), amount of adsorbent (20 mg PP) and acidity of the solution ( $10^{-3}$  M  $\text{HNO}_3$ ) fixed. Similarly, weight variation of the adsorbent was done by varying the amount of PP (5, 20, 40, 60, 80, 100 mg) keeping shaking time (10 min) and acidity of the solution ( $10^{-3}$  M  $\text{HNO}_3$ ) fixed. After centrifugation, 1.5 mL of supernatant was taken out. For every set of experiment, the extent of adsorption by PP was calculated by comparing against reference solution.

## Results and discussions

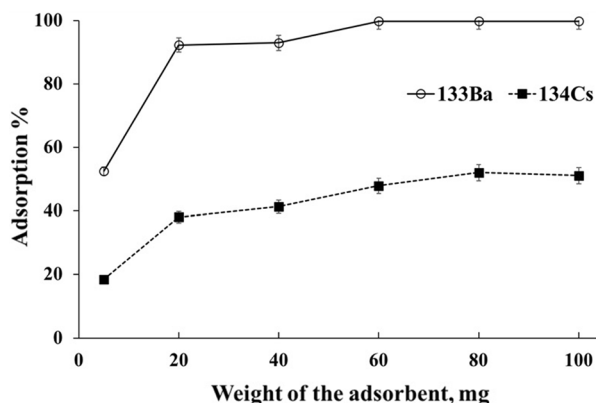
Dried potato peel showed adsorption of both  $^{133}\text{Ba}$  and  $^{134}\text{Cs}$  from the binary solution. Adsorption trend is similar for both the radioisotopes, but adsorption of  $^{133}\text{Ba}$  by PP is higher compared to that of Cs. The adsorption profile of  $^{133}\text{Ba}$  and  $^{134}\text{Cs}$  at different acidic concentrations has been shown in Fig. 1. Maximum adsorption of barium ( $\sim 92\%$ ) was observed at  $10^{-3}$  M  $\text{HNO}_3$  acid concentration followed by steady decrease in adsorption reaching minimum at 0.1 M concentration, followed by slight increase at 1 M  $\text{HNO}_3$  concentration. A steady decrease on adsorption of  $^{134}\text{Cs}$  was observed with increasing acid concentration.

Adsorption of  $^{133}\text{Ba}$  and  $^{134}\text{Cs}$  at different shaking times did not show much variation. Initially adsorption had increased for both  $^{133}\text{Ba}$  and  $^{134}\text{Cs}$  but after 10 min, adsorption process was almost saturated.

Keeping the  $\text{HNO}_3$  concentration fixed at  $10^{-3}$  M and shaking and settling time 10 min, the amount of bio-adsorbent was varied (Fig. 2). Initially the extent of adsorption increased with increasing amount of adsorbent and reached a saturation after 60 mg adsorbent. When 60 mg PP was taken,



**Fig. 1** Adsorption profile of  $^{133}\text{Ba}$  and  $^{134}\text{Cs}$  with varying  $\text{HNO}_3$  concentration keeping bio-adsorbent mass fixed at 20 mg and shaking time 10 min



**Fig. 2** Adsorption profile of  $^{133}\text{Ba}$  and  $^{134}\text{Cs}$  with varying sorbent amount keeping  $\text{HNO}_3$  concentration fixed at  $10^{-3}$  M and shaking time 10 min

the extent of adsorption for  $^{133}\text{Ba}$  was  $\sim 99.8\%$  and that of  $^{134}\text{Cs}$  was  $\sim 48\%$ . With further increase in the amount of sorbent-material (80, 100 mg), uptake of  $^{134}\text{Cs}$  increased to  $\sim 50\%$ . Adsorption (%) and distribution coefficients,  $K_d$  ( $\text{cm}^3 \text{g}^{-1}$ ) of  $^{133}\text{Ba}$  and  $^{134}\text{Cs}$  radionuclides by the sorbent-material at different conditions have been provided in Table 2. The distribution coefficient  $K_d$  of the radioisotopes by the sorbent-material has been calculated from the following equation:

$$K_d (\text{cm}^3 \text{g}^{-1}) = \frac{(\text{Initial activity of Ba-133 or Cs-134}) - (\text{Activity of Ba-133 or Cs-134 after adsorption})}{\text{Activity of Ba-133 or Cs-134 after adsorption}} \times \frac{\text{Volume of the solution (cm}^3\text{)}}{\text{Mass of the adsorbent (g)}}$$

**Table 2** Performance characteristics of potato peel in sorption of  $^{133}\text{Ba}$  and  $^{134}\text{Cs}$  from aqueous solutions

$\text{HNO}_3$ (conc.)	Weight (mg)	V/m ( $\text{cm}^3 \text{g}^{-1}$ )	Ba sorption%	$K_d$ ( $\text{cm}^3 \text{g}^{-1}$ )	Cs sorption%	$K_d$ ( $\text{cm}^3 \text{g}^{-1}$ )
$10^{-4}$	20	200	91.45	2139	48.63	189
$10^{-3}$	20	200	92.32	2404	38.06	123
$10^{-2}$	20	200	55.48	249	17.05	41
$10^{-1}$	20	200	0.01	0.02	7.16	15
1	20	200	5.92	13	13.26	31
$10^{-3}\text{M}$	5	800	52.53	875	18.49	181
$10^{-3}\text{M}$	20	200	92.32	2404	38.06	123
$10^{-3}\text{M}$	40	100	92.97	1322	41.36	71
$10^{-3}\text{M}$	60	67	99.78	30388	47.93	62
$10^{-3}\text{M}$	80	50	99.78	22677	52.07	54
$10^{-3}\text{M}$	100	40	99.78	18142	51.09	42

Similar to Fig. 2,  $K_d$  values decreased for both Ba and Cs radionuclides with increase in acid concentration. With variation in sorbent weight from 5 to 100 mg, V/m ratio varied from 800 to  $40 \text{ cm}^3/\text{g}$ . The maximum  $K_d$  value for Ba ( $3 \times 10^4$ ) was obtained when 60 mg PP was used as sorbent at pH 3 (10 min shaking and settling time). The  $K_d$  value of Cs remained low ( $\sim 10^2$ ) in the entire experiment which indicates a good separation between Ba and Cs radionuclides might be possible.

In Table 3, the present adsorption data has been compared with few reported bio-sorbents used for removal of Ba and Cs from aqueous solutions [32, 35–47].  $K_d$  values obtained

for Ba adsorption by dried potato peel is better when compared to Ba adsorption by *Aloe vera* waste. At best condition,  $K_d$  values for Ba for *Aloe vera* waste, acid treated *Aloe vera* waste and base treated *Aloe vera* waste were 360, 380 and  $1310 \text{ cm}^3 \text{g}^{-1}$  respectively [32]. Also, PP is efficient in terms of  $K_d$  values when compared to that of calcium alginate (CA) beads. At pH = 7, 10 min shaking time, 60 CA beads,  $K_d$  value of Ba was found to be  $86.48 \text{ cm}^3 \text{g}^{-1}$  [27].

The extent of Cs sorption in the present experiment is comparable to that of mucilaginous seed biomass, which was reported to extract 40% of Cs at pH = 6.5 [43]. Examples of some other reported bio-sorbents are edible mushroom

**Table 3** Comparison table providing the optimum conditions of some reported adsorbents used for sorption of Ba and Cs from aqueous solutions

Sorbent-material used	pH, time (min), temperature (K)	$K_d$ ( $\text{cm}^3 \text{g}^{-1}$ ) or adsorption (%)	References
<i>Bio-adsorbents for Ba</i>			
Rice-straw based biochar	8, 15 h, 323	$40 \text{ cm}^3 \text{g}^{-1}$ (97.5%)	Younis et al. [35]
Aloe vera waste	5, 60, 293	$360 \text{ cm}^3 \text{g}^{-1}$	Kapashi et al. [32]
Acid treated <i>Aloe vera</i>		$380 \text{ cm}^3 \text{g}^{-1}$	
Base treated <i>Aloe vera</i>		$1310 \text{ cm}^3 \text{g}^{-1}$	
PVA-alginate beads	8, 120, 298	99%	Majidnia et al. [36]
Dolomite powder	5.5, 120, 293	30%	Ghaemi et al. [37]
Modified ash-based sorbent	4, 40, 323	> 99%	Noli et al. [38]
Potato peel waste	3, 10, 298	99%	Present study
<i>Bio-adsorbents for Cs</i>			
Oxidised bamboo charcoal	12, 180, 288	96%	Khandaker et al. [39]
Modified sugarcane bagasse	10, 180, 288	90–92%	El-Din et al. [40]
Modified persimmon tannin and tea leaves	6.5, 5, 233	> 99% and 80%	Gurung et al. [41]
<i>Punica granatum</i> peel	7, 180, 333	50–60%	Attia et al. [42]
Mucilaginous seeds	6.5	40%	Sangurdekar et al. [43]
Natural and modified tripolite	5.5, 120/30	96–99%	Ivantes et al. [44]
Ti-Ca-Mg based sorbent	9, 180	$4\text{--}13 \cdot 10^5 \text{ cm}^3 \text{g}^{-1}$	Maslova et al. [45]
Porous titanosilicate xerogels	2–12, 10, 298	$8 \cdot 10^5 \text{ cm}^3 \text{g}^{-1}$	Oleksienko et al. [46]
Modified activated charcoal	6.8, 180, 333	93%	Ali et al. [47]
Potato peel waste	3, 10, 298	48%	Present study

(*Pleurotus citrinopileatus* species), which was found to adsorb  $^{137}\text{Cs}$  [48]; common yeast, *Saccharomyces cerevisiae*, which could selectively uptake long-lived  $^{152,154}\text{Eu}$  from a mixture of  $^{152,154}\text{Eu}$ ,  $^{60}\text{Co}$  and  $^{134}\text{Cs}$  without accumulation of  $^{60}\text{Co}$  and  $^{134}\text{Cs}$  [49]; marine cyanobacteria for Cs(I) sorption as reported by Yu et al. [50], etc.

The absorption mechanism onto bio-sorbents is a complex process and multiple mechanism like electrostatic interaction, ion exchange or adduct formation with the surface functional groups may work together. For the present experiment, a careful observation to the Fig. 1 may vouch against the electrostatic attraction. A point of zero charge (PZC) is observed in case of electrostatic attraction. It is the point beyond which no metal-adsorbent interaction occurs. From Fig. 1, it is clear that there is no PZC for Cs. Though at 0.1 M concentration Ba adsorption is zero, but beyond 0.1 M adsorption starts again, which should not be the case for electrostatic attraction.

The adsorption process observed in the present experiment can be explained by the ion exchange mechanism, where both  $\text{Ba}^{2+}$  and  $\text{Cs}^+$  are in competition for the binding sites. pH is one of the significant factors affecting the adsorption process. Due to the higher charge density of  $\text{Ba}^{2+}$ , it easily replaces  $\text{H}^+$  from protonated PP surface as compared to  $\text{Cs}^+$  and adsorption of  $^{133}\text{Ba}$  is more than that of  $^{134}\text{Cs}$  under all experimental conditions. According to Tran et al. [51], bio-sorbents exhibit a low specific surface area and total pore volume but have abundant surface functional groups, and a high net surface charge density. We have analysed dried PP under FTIR spectroscopy. Analysis of the sorbent-material showed a strong peak at  $3332\text{ cm}^{-1}$ , which confirms the presence of H-bonded  $-\text{OH}$  stretching. The hydroxyl group might belong to carboxylic acid, phenol or alcohols, which are present within lignin, hemicellulose and cellulose moieties of the adsorbent.  $-\text{C}=\text{O}$  stretching of aldehydes and ketones was observed at  $1591\text{ cm}^{-1}$ . Potato peels generally consist of lignocellulosic residues comprising of cellulose, hemicellulose and lignin. These components possess different functional groups such as phenolic, hydroxyl, carboxylic, amines and ether groups, which can also contribute to physical and chemical interactions via surface complexation, adduct formation or hydrogen bonding [52, 53]. The adsorbent is rich with oxygenated surface groups, which can easily attract and bind cations like  $\text{Ba}^{2+}$  or  $\text{Cs}^+$ . Bivalent  $\text{Ba}^{2+}$  naturally is more efficient for these interactions.

## Conclusions

Some thrown away agro-wastes have already been established as adsorbent for long-lived radionuclides. Earlier, we have studied the efficacy of potato peel towards adsorption

of  $^{88}\text{Zr}$  radioisotope [54, 55]. The present experiment successfully added potato peel (*Solanum tuberosum*) to the list as adsorbent of  $^{134,137}\text{Cs}$  and  $^{133,140}\text{Ba}$  radionuclides. At the optimum conditions of  $\text{pH}=3$ , 60 mg adsorbent, 10 min shaking time and 298 K, maximum  $>99\%$  of  $^{133}\text{Ba}$  was adsorbed by dried potato peel.  $K_d$  ( $\text{cm}^3\text{ g}^{-1}$ ) value calculated at this point was  $30,388\text{ cm}^3\text{ g}^{-1}$ . Under the same condition, maximum 48% of  $^{134}\text{Cs}$  was adsorbed by dried potato peel. However, poor  $K_d$  ( $\text{cm}^3\text{ g}^{-1}$ ) values were obtained for  $^{134}\text{Cs}$ . It is true that the maximum adsorption for  $^{134}\text{Cs}$  was  $\sim 50\%$  in the present experimental condition. But multiple extraction by potato peel would increase the extent of extraction. For example, three times extraction would lead to about 87% extraction of caesium radionuclides. Hence, this study is an attempt to investigate the efficacy of potato peel as a bio-adsorbent for Ba and Cs radionuclides. However, bio-sorbents, by definition are bio-degradable and is not compatible with inorganic matrices with respect to cementation and classing processes for final deposition of radionuclides.

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## Compliance with ethical standards

**Conflict of interest** The authors declare that they have no conflict of interest.

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