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Radiological dose assessment of naturally occurring radioactive materials generated by the petroleum industry in wildlife: A case study of chinkaras of Lavan Island, Iran

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Original Article

Abstract

Human activities such as oil and gas production can enhance the natural level of naturally occurring radioactive materials (NORM) in by-product and waste streams. Iran has been among the top five oil producing countries since 2005. This high production rate emphasizes the importance of NORM management to ensure the safety of humans and wildlife. Petroleum storage and transport facilities are located at Lavan Island, Iran. Presence of animals including dolphins, sea turtles, and chinkaras make this island one of the most unique wildlife refuges in Iran. This paper combines waste disposal methods relevant to the petroleum offshore industries, NORM waste characteristics, and geographical, geological, and climate conditions of Lavan Island in order to develop enveloping exposure scenarios. Sludge burning is determined as the most concerning scenario by assuming chinkaras as the endpoint. Ecological and radiological assessment procedure is modeled with MATLAB-Simulink as a dynamic system. Clearance level for radiation protection of chinkaras is calculated as 41 Bq/kg. This value may be insufficient for radiation protection of workers, because exposure pathways are not derived based on human behavior. According to environmental pathways and condition of chinkaras, this value sufficiently covers all aspects of radiation protection.

KEYWORDS: Radioactive Wastes, Radiological Health, Radioactive Soil Pollutants, Radioactive Food Contamination, Radiation Protections

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Introduction

The majority of radionuclides in naturally occurring radioactive materials (NORM) are essential constituents of the earth's crust.[1] Primordial radionuclides ^{238}U, ^{232}Th, and ^{235}U, are the parent radionuclides for the three naturally occurring decay chains commonly called the uranium, thorium, and actinium series, respectively.[2]

There are many industries and human activities that can enhance the level of NORM in their waste streams and by-products. It should be noted that NORM is typically associated with non-nuclear industries.[3] Regardless of the activity from which NORM originates, there are six types of materials which could contain the elevated levels of radiation; waste rock, sand slag ash, sludge, and scale. Oil and gas production and processing is one of the most

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notable industries that produces NORM, mainly in the form of sludge and scale.[4]

Uranium and thorium series exist in various concentrations in unground formations. Both ^{238}U and ^{232}Th are relatively insoluble and remain in place in subsurface formations; however, some of their decay products [^{226}Ra (from uranium series) and ^{228}Ra (from thorium decay series)] are slightly soluble and can become mobilized in liquid phase of the formation.[5]

During drilling and production operations, produced water is extracted from underground formations and brought to the surface. Discharge of the produced water into the marine environment is a typical disposal method in Brazil[6], European commission,[7] and many other countries. If the produced water is disposed in surface pits, the liquid content would evaporate, and thus, the concentration of NORM in the pits could be considerably enhanced.

Many studies have investigated the radiological hazards of NORM in the petroleum industry[8-10] and a limited number of researches have been conducted on NORM originated in the Iranian petroleum industry[11,12] which prove the existence of NORM in scales and sludges.

Gas oil separation plants (GOSP), storage tanks, and pipelines are possible locations for accumulation of sludge and scale. In Iran, scales are generally discharged into the sea, surface ponds, or landfills along with other kinds of wastes. Other methods, like encapsulation, which are widely used around the world[5], are not common in Iran. On Lavan Island, extracted scales are contained and stored in drums. Scales which are extracted offshore are usually discharged into the water. None of the wastes are landfilled on the island, so related exposure pathways are excluded from radiological assessment.

Incineration, land spreading, and landfilling are common practices applied for sludge management in Iran. These methods could harm both human and environmental health, if done without any appropriate system and monitoring. Sometimes, on Lavan Island, most of the sludges are directly or accidentally burnt in the outdoor environment without any suitable isolation.

The main waste generators on Lavan Island are the Iranian Offshore Oil Company (IOOC), Lavan Oil Refining Company, Lez village, military facilities, and other sources including domestics.

On the island, there are facilities to contain and transport crude oil which produce significant amounts of sludge. In case of leakage, sludge can be burnt or contained in metallic drums. These drums are transported back to the onshore facilities and will not be included in the Lavan Island radiological assessment. Sludge burning is the only significant disposal scenario concerning chinkaras on the island. It should be noted that sludge burning is not a standard waste disposal activity, but sometimes it is unavoidable.

There are approximately 400 chinkaras on the Island. Due to absence of natural predators, lack of food, and limited pastures, most of them are attracted to residences and waste storage facilities. These facilities contain NORM; hence, radioactive exposure is inevitable. Sludge burning, and thus, ash settlement on the pastures is an important exposure pathway, which has been investigated thoroughly in this paper.

Materials and Methods

This research has been conducted by following "the principles of radiological ecological risk assessment"[13]. This study attempts to develop real and pervasive scenarios according to personal visits and the International Atomic Energy Agency (IAEA) context[14], in order to predict the annual dose of NORM received by chinkaras. Each model consists of three major parts; source term, environmental pathway, and exposure pathway. Source term demonstrates the radioactive decay and growth inside the waste package. Environmental pathway investigates the transport of radionuclides from waste to a defined endpoint. In this paper, chinkaras are chosen as the endpoint. Exposure pathway encompasses possible ways through which radionuclides may come into contact with the endpoint.

Incineration without any protection and equipment is not a favorable option. Open burning produces plume. If the plume moves toward inland, the following exposure pathways would be possible regarding chinkaras as the endpoint.

- Submersion in plume: Being submersed in the plume could expose the creature to NORM. The main pathways would be external irradiation and inhalation of NORM particles.
- External irradiation from soil: After deposition of plume on the surface of soil, external exposure from NORM particles could be a significant pathway. Due to lack of surface water and even fresh water ponds on the island, external irradiation from water is negligible.

Ingestion of pasture vegetation, soil, and water: Chinkaras graze in the limited meadows of the island every day. If NORM particles are deposited on the surface of pasture, soil, or water, chinkaras will ingest them without noticing the contamination. The conceptual model of the sludge burning scenario is illustrated in figure 1.

This paper assigns the local data of Lavan Island into the ecological and radiological dose assessment models. Most mathematical relations and equations are taken from the Improvement of Safety Assessment Methodologies for Near Surface Disposal Facilities (ISAM) project.[15] Generic models have been applied in this study. Generic models are the best tools to derive standard limits due to their reasonable degree of conservatism and reliable results. These results can be used as the basic standards for protection of chinkaras against NORM incineration accidents.

Source Term Modeling

The following differential equation has been used to model NORM decay chains in this paper[16]:

$$\frac{dN_i(t)}{dt} = \lambda_{i-1}N_{i-1}(t) - \lambda_i N_i(t)$$

where N_i and λ_i are the quantity and decay constants of i^{th} nuclide, respectively.

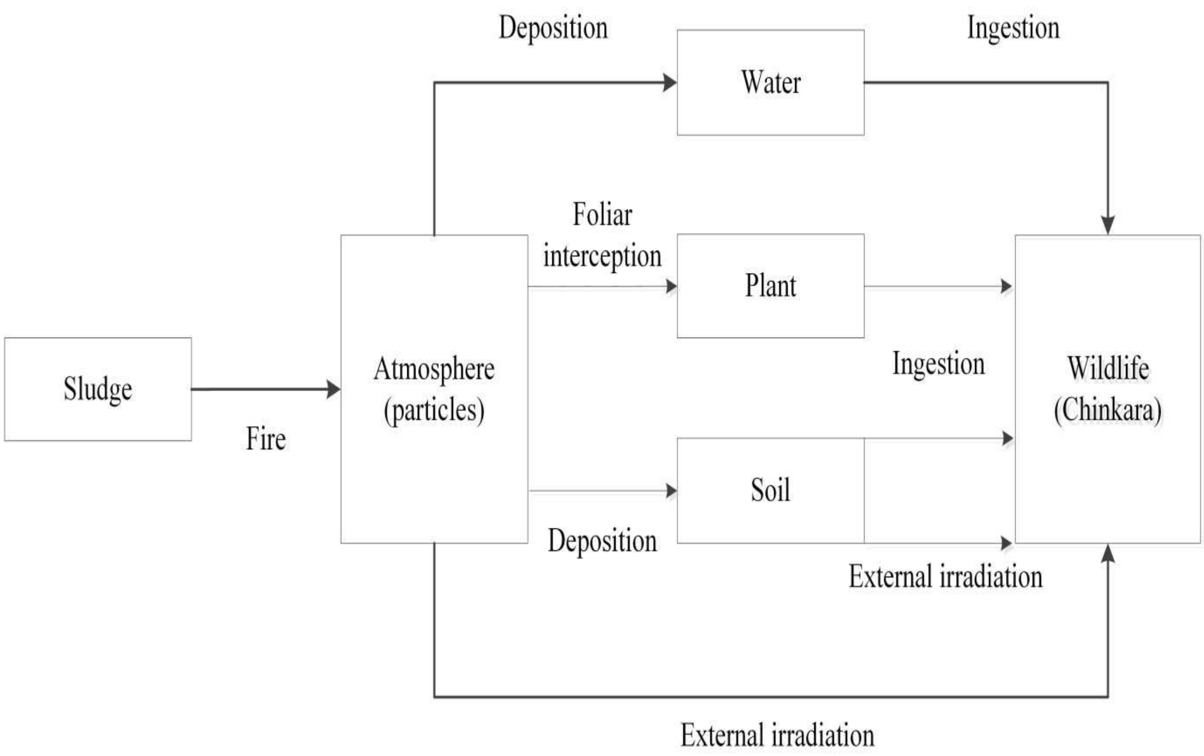

Figure 1. Conceptual assessment model of sludge burning scenario

Environmental Pathway Modeling

The release rate of a radionuclide from the fire, R_{fire} (Bq/h), is obtained through the following equation:

$$R_{fire} = f_{rel,fire} A_m V_{fire} \rho_{bd} / t_{fire}$$

where $f_{rel,fire}$ is the release fraction for radionuclide I (unitless), A_m is the specific activity of the radionuclide in the sludge (Bq/kg), V_{fire} is the volume of the waste consumed in the fire (m^3), ρ_{bd} is the bulk density of the waste (kg/m^3), and t_{fire} is the duration of the fire (h).

It is cautiously assumed that the associated plume is neutrally buoyant. Air concentration of a radionuclide at ground level, $C_{air,fire}$, is obtained by:

$$C_{air,fire} = R_{fire} C_{integ.fire}$$

where $C_{integ,fire}$ is the time-integrated air concentration at ground level [(Bq/m^3) per (Bq/h)]. The above equation cautiously assumes that the plume is non-depleting during passage towards the endpoint (each individual chinkara).

The surface concentration of a radionuclide, $C_{surf,fire}$, resulting from deposition is:

$$C_{surf,fire} = C_{air,fire} t_{dep,fire} (V_{g,fire} + W_{out,fire} h_{fire})$$

where $t_{dep,fire}$ is the time over which deposition occurs (s), $V_{g,fire}$ is the dry deposition velocity (m/s), $W_{out,fire}$ is the washout coefficient (1/s), and h_{fire} is the plume height (m).

Exposure Pathway Modeling

The total dose to an individual chinkara living on the island can be expressed as:

$$Dose_{total} = Dose_{sub} + Dose_{ext} + Dose_{ing}$$

where $Dose_{sub}$ is the dose due to the external irradiation from submersion in the plume (Sv/y), $Dose_{inh}$ is the dose from inhalation of NORM particles (Sv/y), $Dose_{ext}$ is the dose from external irradiation of deposited NORM on the ground (Sv/y), and $Dose_{ing}$ is the dose from the ingestion of NORM deposited on the pasture (Sv/y).

Dose of external irradiation from submersion in the plume can be obtained by:

$$Dose_{sub} = C_{air,fire} t_{e,out} DF_{sub} Occ_{fire}$$

where $t_{out,fire}$ is the exposure time to one occurrence of fire (h), DF_{sub} is the dose factor for external irradiation from submersion in the plume [(Sv/h) per (Bq/m^3)], and Occ_{fire} is the number of fires per year (1/y).

External dose by direct irradiation from soil can be expressed as:

$$Dose_{ext,soil} = C_{surf,fire} t_{e,out} DF_{ext,soil}$$

where $DF_{ext,soil}$ is the dose coefficient for contaminated soil ((Sv/h) per (Bq/m^2)).

Chinkaras may receive internal radiation doses after ingesting contaminated pasture vegetation, soil, or water. Internal dose according to ingestion of contaminated material can be expressed as:

$$Dose_{ing} = \sum_{i=pasture,soil,water} CN_{surf,fire} Q_i Occ_{fire} t_{pst} \left[\frac{(1 - e^{-\lambda_{pst} t_{pst}})}{\lambda_{pst}}\right] DF_{ing}$$

where $CN_{surf,fire}$ is the activity concentration of radionuclides on the unit surface of the edible materials (food) ((Bq/kg) per (Bq/m^2)), Q_{pst}, Q_{soil}, and Q_{water} are the pasture vegetation, soil, and water consumption rate (kg/d), respectively, λ_{pst} is the effective rate constant for removal of activity from the pasture (1/d), t_{pst} is the time following the release over which the pasture vegetation are consumed (d), and DF_{ing} is the dose conversion factor for ingestion (Sv/Bq). It should be mentioned that sludge burning has been considered as an accidental phenomena.

Parameters regarding to waste characteristics and chinkaras' habitat and diet are gathered from local people and organizations. Parameters for radionuclide transfer coefficients and other radiological and biological factors are adapted from relevant studies.[17-21]

The dose criterion of 1 mSv/yr is considered to fulfill radiation protection of the public and workers. In this paper, the same limit is assigned as annual dose limit for total exposures of chinkaras to NORM sources.

Mathematical relations which were used in this paper are modeled with MATLAB-Simulink (The MathWorks Inc., Natick, MA, USA). The MathWorks MATLAB software and its associated graphical extension Simulink have the ability to simulate dynamic systems with ease and flexibility.[22] Simulink is very effective in the analysis of the time variable signals and systems. In this research, fate and transport (contaminant migration through the environment) of NORM are considered as a dynamic system which varies with time; therefore, it has been modeled with Simulink. In addition to providing a user friendly environment, any modification and upgrade to the initial model can be easily performed using Simulink features.

Results and Discussion

NORM residue properties are based on previously conducted researches in Iran.[11,12] Moreover, meteorological data were taken from local agencies. Table 1 indicates source term characteristics.

Exposure pathway parameters were broadly taken from personal observations, local agencies, and general assumptions for near surface disposal facilities.[14] There is no documented or official record about the chinkaras' diet and habitat. In order to gain the most reliable results, these data were gathered from local veterinarians and the Health, Safety, and Environment (HSE) offices. Exposure related parameters are illustrated in table 2. Exposure dose coefficients, which include the ingestion, inhalation, and external irradiation pathways, are broadly consistent with the database of radionuclide transfer parameters for freshwater wildlife.[24]

Occurrence of fire is assumed as an accident and is not very probable due to the safety protocols and equipment installed at the field. This paper takes into account the worst case scenario which investigates an accident that may occur once a year. Due to the large volume of sludge, each accident is expected to last for a maximum duration of 12 hours. The HSE and fire department of petroleum facilities are capable of extinguishing the fire in a few hours, but in this case, the most conservative and realistic values are considered. Local interviews and surveys play the key role in gathering of these data.

Chinkaras' diet usually consists of natural vegetation of the island and the food provided by private contractors. They might also search for food in garbage bins, but this would not be considered as their common food source. In addition to the pasture vegetation and water, they consume a small proportion of soil with vegetation. This soil might be ingested via attachment to the roots or deposition of dust on the vegetation. Although the quantity of consumed food depends on many factors, like the gender and age of the animals, a typical fully grown male is taken into account in this research. Chinkaras' diet, behavior, and habitat pattern are also determined by local surveys and information from the local authorities. λpst, which indicates the removal of activity from the pasture, is directly adopted from IAEA documents.[14]

These parameters are considered to be the most conservative data in order to fulfill the maximum protection goals. Calculations are carried out mainly for ^{226}Ra and its daughters due to their presence in sludge. By applying the data from tables 1 and 2 to the exposure model, figure 2 was achieved.

Table 1. Source term characteristics

$f_{rel,fire}$ (-)	A_m* (Bq/kg)	m (T)	V_{fire} (m^3)	ρ_{bd} (kg/m^3)	t_{fire} (h)	$C_{integ,fire}$** (Bq/m3) per (Bq/h)	$t_{dep,fire}$ (s)	$V_{g,fire}$ (m/s)	$W_{out,fire}$ (1/s)	h_{fire} (m)
0.001	Ra: 25.7 Pb: 25.7 Po: 25.7	1	1.8 E+7	1800	6	3.24	3600	0.02	0.003	12

*Radium and its progenies have been considered to be in secular equilibrium due to long retention time in storage tanks before disposal.
** Time-integrated air concentration at ground level is based on dominant atmospheric conditions of Lavan Island.[23]

Table 2. Exposure term characteristics for an individual chinkara

$t_{out,fire}$ (h)	Occ_{fire} (1/y)	Q_{pst} (kg/d)	Q_{soil} (kg/d)	Q_{water} (kg/d)	λ_{pst} (1/d)	t_{pst} (d)
12	1	2.1	0.1	0.3	0.02	0.5

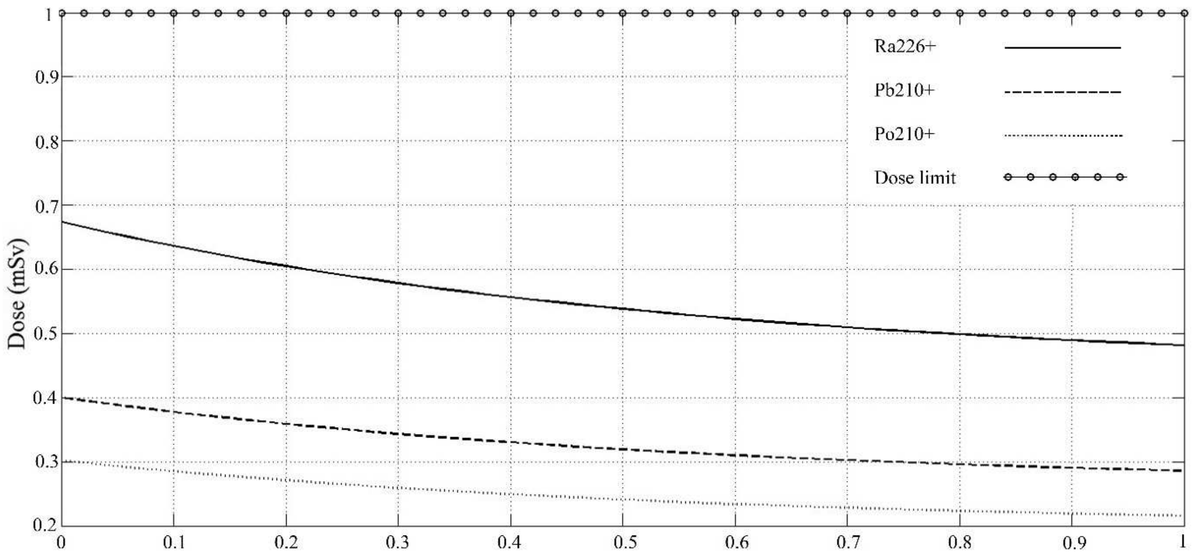

Figure 2. Dose assessment results for the chinkaras

It can be concluded that the wastes sampled at Lavan Island are cleared from any radiological hazards due to their lower dose responses compared with the standard limit. Figure 2 indicates an exponential trend of data. This is due to the decay of radium and its progenies in the environment. The longer it takes for the chinkaras to consume the pasture vegetation, the lower the dose they will, probably, receive. It is not easily possible to prevent the chinkaras from grazing in contaminated areas. If a chinkara preservation program is designed, their pasture should be carefully isolated in case of NORM contamination or completely replaced by non-contaminated food.

In order to calculate the accurate cleared values, iteration approach and reverse analysis should be applied to initial activity concentration and the results continuously compared with dose limit. The model has been executed 70 times until the initial activity concentration satisfied the dose criteria. The results were continuously examined and modified to converge to a singular value. By following the aforementioned procedure, clearance levels for each radionuclide have been calculated and illustrated in table 3.

Table 3. Proposed clearance levels for sludge burning scenario

Radionuclide	Activity concentration (Bq/kg)
$^{226}Ra^{*}$	41
$^{210}Pb^{*}$	70
$^{210}Po^{*}$	92

* Also includes daughters

Each of these values illustrates the maximum concentration for each radionuclide that causes no harm to chinkaras after an accidental fire. In other words, the minimum activity concentration of ^{226}Ra+, ^{210}Pb+, and ^{210}Po+ should be 41, 70, and 92 (Bq/kg), respectively, in order to initiate the chinkara food preservation program. Considering the importance of radiation protection and conservative assumptions, the minimum value of clearance levels is usually declared as the general clearance level. In the present study, 41 Bq/kg (activity concentration of ^{226}Ra+) was chosen as the activity limit for sludge burning scenario.

This implies that if the activity concentration of sludge is higher than 41 (Bq/kg) and chinkaras are exposed to fire, their' food and habitat should be isolated or decontaminated based on domestic policies. These efforts will ensure the protection of chinkaras from radiation hazards of NORM.

This value is not suitable for radiation protection of workers, because exposure pathways are not defined due to human behavior. According to environmental pathways and condition of chinkaras, this value sufficiently covers all aspects of ecological radiation protection. If humans are considered as the endpoint, new sets of scenarios must be developed.

This scenario covers accidental fire situations, but it could be applied to open burning of sludge. Although open incineration is not a recommended and safe disposal method, it should be considered that, in the case of open incineration, the activity of NORM should not exceed the clearance level.

In addition to sludge, hard scales from pipelines should also be investigated thoroughly. Activity concentration of scales has been recorded as the highest among other NORM wastes generated in the petroleum industry. Due to the discharge of scales into the Persian Gulf and their absence on the Island, their radiological assessment falls outside the scope of this study.

Conclusion

Typical measurements around the world indicate the average activity of sludge to be between 50 and 800000 Bq/kg. The only official document regarding NORM activity in the Iranian petroleum industry indicates the activity range of 10.6 to 1480 Bq/kg for ^{226}Ra+. This research recommends the maximum activity limit of 41 Bq/kg for sludge wastes in Lavan Island which satisfies the radiological protection criteria for the chinkaras. Any waste with lower activity is simply cleared from further protection policies, but in case of higher activity, appropriate disposal and protection are required.

Conflict of Interests

Authors have no conflict of interests.

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