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A New Approach to Derive Clearance Levels for Wastes Containing Naturally Occurring Radioactive Materials (NORM) (Case Study: Lavan Island, Iran)

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ABSTRACT

Naturally Occurring Radioactive Materials (NORM) are present in earth's crust with different concentrations. Human activities such as oil and gas production and mineral extraction and processing could enhance the natural level of NORM in by-products and waste streams. To protect public and workers from these enhanced NORM sources, clearance levels should be established. Clearance levels are regulatory limits which are defined in order to protect the environment and human from radiation sources. This study proposes a new approach to derive both generic and specific clearance levels. This approach consists of two scenario systems; Generic Scenario System (GSS) and Specific Scenario System (SSS). GSS is a comprehensive assessment tool which can be applied for any country. GSS is assembled by extensive research and actual NORM management methods. GSS investigates the radiological impacts of NORM from generation through final disposal. SSS is acquired through combination of GSS with field data and observation. This approach simultaneously considers both human health and economic aspects. To examine the implication of this method, Lavan Island in Persian Gulf is chosen for case study. The comparison between the generic and specific approaches for Lavan Island indicates that most exposure scenarios in generic approach which could result in extremely conservative values do not exist in specific scenarios regarding Lavan Island. Exposure groups, waste management options and enclosed circumstances of Islands, make them a preferable location to distinguish between SSS and GSS and understand the approach of this paper.

INTRODUCTION

NORM: Naturally Occurring Radioactive Materials (NORM), which are enhanced by industrial process, are also referred as TENORM (Technologically Enhanced Naturally Occurring Radioactive Material) in researches and literatures. In this paper they are both simply called as NORM. Naturally occurring radionuclides such as the ones from the ^{238}U and ^{232}Th series, as well as ^{40}K, are essential constituents of the earth's crust and can be found in higher than normal concentrations in some regions of the planet, known as "high natural radiation areas" (Gazineu & Hazin 2008).

The majority of radionuclides in NORM (principally radium and radon) arise from uranium and thorium decay. Primordial radionuclides ^{238}U, ^{232}Th and ^{235}U are the parent radionuclides for the three naturally occurring decay series commonly called the uranium, thorium and actinium series, respectively (Ojovan & Lee 2005). Occurrence of actinium series is extremely rare in nature which makes its radionuclides negligible NORM components especially in radiological assessments.

NORM activities could be significantly enhanced in waste stream and by-product of some human activities. Physical and chemical procedures like grinding, incineration, smelting, leaching etc., which usually alter the matrix and molecular bonding of materials, are responsible for the enhancement of activity levels in wastes or by-products. A considerable body of knowledge and experience has already been built up concerning operations involving minerals and raw materials (in addition to uranium ores) that may lead to a significant increase in exposure to natural sources. The following industries have been sorted in descending order of priority for radiological protection requirements (IAEA 2006): extraction of rare earth elements; production and use of thorium and its compounds; production of niobium and ferro-niobium; mining of ores other than uranium ore; production of oil and gas; manufacture of titanium dioxide pigments; the phosphate industry; the zircon and zirconia industries; production of tin, copper, aluminium, zinc, lead, and iron and steel; combustion of coal and water treatment.

TENORM is typically associated with non-nuclear industries. Uranium milling tailings (UMT) are a component of the nuclear fuel cycle, and have generally been considered distinct from TENORM (Landa 2007). Regardless of the activity which NORM originates from, there are six types of materials which could contain the elevated levels of radiation: waste rock; sand; slag; ash; sludge and scale.

Other material types like used filters, scrap metals and produced fluids could also have enhanced or considerable amounts of NORM. These materials are related to specific industries and should be analysed in specific radiological assessments. This papers deals with petroleum industry and introduces a new approach to drive both generic and specific clearance levels for NORM.

NORM in petroleum industry: Iranian petroleum industry is divided into two main sectors: Upstream and downstream. Upstream sector which widely called E&P (Exploration and Production) includes the exploration for potential oil and gas reservoirs, drilling of exploratory wells and subsequently production wells that recover and bring the crude oil and/or raw natural gas to the surface. The downstream sector involves the refining and processing of hydrocarbons into usable products such as gasoline and variety of petrochemical products. The main producer of NORM in petroleum industry is upstream sector. Two out of six NORM categories and one particular type exists in petroleum industries which are scale, sludge and produced water respectively.

Uranium and thorium series exists 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 are slightly soluble and can become mobilized in liquid phase of the formation (Smith et al. 1996). The soluble progenies include ^{226}Ra and its daughters ^{222}Rn and ^{210}Pb (decay products of uranium series) and ^{228}Ra (from thorium decay chain).

Underground oil and gas reservoirs are usually accompanied by large amount of water which is widely mentioned as produced water. Produced water is highly saline and contains high concentration of chloride and low concentration of sulfide compounds (due to reduction condition in underground environments). During drilling and production operations, produced water is extracted from underground formations and brought to surface which contains considerable amounts of ^{226}Ra and ^{228} Ra. For gas reservoirs ^{222}Rn migrates with gas to surface.

The reservoir's fluids are directly sent to GOSP (gas-oil separation plant) in order to separate gas, oil and water and then store them in different tanks. GOSP, storage tanks and pipelines, which could contain sludge and scale, are the primary sources of NORM in production facilities.

Injecting produced water to deep subsurface structures and EOR (enhanced oil recovery) wells, discharging to evaporation ponds or simply to surrounding marine environment are basic disposal options that are largely used by companies in Iran.

Solubility of radium isotopes in water depends mainly on pH and salinity of water. By increasing salinity and acidic or basic properties of water, more radium would dissolve in water. On the other hand due to decreasing temperature and pressure, the precipitation rate would increase. Pressure and temperature would drop when produced water is brought to surface and moves inside pipelines. This process would lead to the formation of a hard material known as scale.

Scales could directly affect petroleum rate production by blocking the interior space of pipes. Scales should be removed, hence their extracting, handling and disposal require radiological safety assessments and specific clearance levels. Both mechanical and chemical methods are available to remove scales from pipes or other equipment. In Iran scales are directly discharged in sea or disposed along other kinds of wastes into landfills. Other methods like encapsulation, which is used around the world (Smith et al. 1996), are not very common practices in Iran.

Unlike scales which are produced from water, sludges tend to deposit from crude oil. Heavy oils have more potential to settle their deposits and form sludges. GOSP and oil storage tanks are primary places for sludge formation. Incinerating, land spreading and land filling are common practices in Iran which should be evaluated under radiological safety assessment.

Radon is a radioactive gas which moves with natural gas to the well head facilities. Radon progenies attach themselves to airborne particles and forming thin radioactive layers inside gas processing equipment. These equipment are contaminated with NORM and due to difficulties in removing radioactive film, they should be either recycled or disposed. Gas compressor, scrubbers and pipes are main places which NORM could accumulate.

A limited number of researches have been conducted on NORM originated in Iranian petroleum industry (Khodashenas et al. 2012, Moatar et al. 2009). These researches indicate that water disposal pits should be designed in order to minimize radiological effects both to the people and the environment. According to these studies reinjection of produced water into abandon wells of Iranian Offshore Oil Company in Persian Gulf, have preference over discharging to the pits.

In addition to common six waste types (waste rock, sand, slag, sludge, scale, ash) which are assessed in (EC 2001), water containing NORM is also considered in this study. Produced water from oil and gas industry, wastewater from mining operations and leachate from tailing dams are concerning issues in environmental contamination management. If water is exposed to evaporation, concentration of NORM in liquid phase could be considerably enhanced. This problem is more substantial in arid environments like Lavan Island. These issues are convincing enough to consider liquids as a separate waste type in radiological assessment of NORM.

Lavan island: Lavan is the third big island in the Persian Gulf following Qeshm and KishIslands. Lavan is a populated island with area of 76 km^2 and population of 3000 native individuals. High temperature (50°C) and extremely humid climate are at their apex in summer. Local people are either pearling or working at petroleum facilities. Fig. 1 indicates the location of Lavan Island in Persian Gulf.

There are three major islands in Persian Gulf that each of them is an endpoint for related oil and gas reservoirs. Aforementioned islands are: Lavan, Khark and Siri. Depending on area and volume of underground reservoirs, some production platforms have been established. According to personal observations and documents of Iranian Offshore Oil Company (IOOC), Lavan Island is selected as case study for this paper. At present, Lavan Island has more than 3100 inhabitants, generating up to 7514 kg waste per day. In general, the solid waste generators in Lavan Island are the IOOC, Lavan Oil Refining Company, Lez village, military facilities and other sources including domestics (Shams Fallah et al. 2013). Lavan Island is the centre of four oil and gas reservoirs. Group one (Salman, Reshadat, Resalat and Belal) and group two (Lavan) are oil and gas reservoirs respectively (IOOC 2013).

Discharge of produced water into the marine environment is a typical disposal method in Brazil (Jerez Vegueria et al. 2002), Europe (Betti et al. 2004) and many other countries. On offshore production platforms, most produced water is directly discharged to the Persian Gulf and crude oil and natural gas are sent to the processing facilities located on Lavan Island, while a significant portion of produced water is brought to the island. Scales are formed in pipelines which are extracted by mechanical means (known as pig running operation). Amount of participation of scales is a function of production rate and the time which liquids are still in the pipes. Extracted scales are contained and stored in drums. Scales which are extracted on the platform are usually discharged into the water. No wastes are considered to be landfilled on the island so related exposure pathways would be omitted from radiological assessment.

On the island, there are facilities to contain crude oil until oil tankers arrive for loading. Large volumes of sludge are settled on the bottom of storage tanks. Parts of these sludges are transported to underground trenches which are excavated beneath the tanks. In case of storage tanks leakage, sludges are burnt or contained in the drums. Sludge and scale drum are transported back to the onshore facilities like Bahregan. Shipped sludge and scale drums are not considered in Lavan radiological safety assessment.

Clearance levels: The decay scheme emits high-energy alpha particles at each stage: $^{238}U \rightarrow ^{234}Th \rightarrow ^{230}Th \rightarrow ^{226}Ra \rightarrow ^{222}Rn \rightarrow ^{218}Po \rightarrow ^{214}Po \rightarrow ^{210}Pb$, and finally to stable ^{206}Pb. Radiation causes a wide variety of cancers, including kidney plus liver failure, and lung plus skin cancers (Ernst 2012). Any radiological assessment should be conducted to achieve maximum protection and final results would be known as clearance levels.

Clearance levels are regulatory limits which are defined in order to protect the environment and human from radiation sources. This means that the activity concentration of any radionuclide (either natural or artificial) in waste package must be either equal or lower than these limits. Clearance level calculations are based on the evaluation of a selected set of typical exposure scenarios for all material, encompassing external irradiation, dust inhalation and ingestion (direct and indirect). The values selected were the lowest values obtained from the scenarios (IAEA 2004a).

Regardless of groups, which are coming into contact with NORM (worker or public), two types of clearance levels have been proposed in this paper. Generic clearance levels are the context of regulatory surveillance for macro environmental assessment and specific clearance levels investigate any micro assessment associated with specific industries. Generic clearance levels are reliable tools to distinguish between the radioactive and cleared wastes. There are vast types of wastes which contains NORM; so enveloping scenarios should cover every aspect of radiation exposure. This paper applies waste management elements with scenario development procedure to define a new set of scenarios regardless of waste types. Due to generic nature of assessment, all parameters should be selected with adequate degree of conservatism.

Each industrial branch has its own set of characteristics such as material characteristics (especially activity level), material quantity and preferred waste management option (EC 2000a). Another type of activity limit should be defined for specific activities and industries. These are called specific clearance levels. Derivation of these value requires more degree of realism in selection of parameters and developing most likely scenarios. The reason that makes derivation of specific clearance levels a necessity is to avoid any over estimation of dose to people.

Extensive research and projects have been conducted on derivation of activity limits (clearance levels) for various types of radioactive wastes and waste management op-

Fig. 1: Lavan Island located in Persian Gulf (Google Earth 2013).

tions and facilities (IAEA 2003, IAEA 2004b, EC 2000b, EC 2001, IAEA 2005). Different countries and states prepared their own guidelines for management of NORM in their industries (HC 2000, Arpansa 2008, OGP 2008, Shwd 2011, CAPP 2000). Currently, modelling and calculation are being carried out to define generic and specific clearance levels of NORM in Iran (Sedighian 2013). This paper introduces a new approach to define both generic and specific clearance levels simultaneously.

MATERIALS AND METHODS

Wastes containing NORM could be generated under different conditions. Each industry and process has its own wastes with unique physical and chemical properties. Generic clearance levels should cover all industries and waste streams. In order to achieve this level of confidence, an integrated system of scenarios is developed. This system, referred as Generic Scenario System (GSS), is completely independent of NORM generation process and only focuses on waste categories. Generic assessment would be calculated by assessing GSS and fulfilling these four steps:

1. Identification and characterization of source terms (7 source terms, each with unique physical and chemical properties (density, porosity, leaching rate, etc.).
2. Investigation of possible environmental pathways (surface and ground water, atmosphere, soil and food chain are major environmental pathways).
3. Investigation of exposure pathways (ingestion, inhalation, external irradiation and skin contamination).
4. Determination of endpoint. Endpoint would be specified due to assessment objectives. Human (public or worker), animals or even plants might be considered as endpoints.

Basically, these steps may seem common in most radioactive waste assessments. The main difference is that NORM does not need to be handled under strict conditions and regulations as radioactive wastes in most countries, so without any protective measures and regulations exposure scenarios are more likely to happen. By combining source terms, environmental and exposure pathways and endpoints, GSS would be made. In this paper GSS are developed with regard to waste management operational functions which are shown in Fig. 2. GSS which is applicable for all NORM wastes consists of:

1. Generation
2. Handling and storage
3. Transportation
4. Recycle-Reuse-Recovery (house, playground, road, dam)
5. Controlled disposal (landfilling)
6. Uncontrolled disposal (open dumping, open burning, discharging to water body)
7. Human intrusion (excavation, site dwelling)

GSS should be calibrated with local data of country, state or any desired region. In addition to GSS and local data, the final model should be evaluated with a dose criterion. The article 31 experts propose to set the criteria for clearance-exemption for work activities at an annual effective dose increment of 300 μSv (EC 2001). This can be converted in terms of activity concentrations (clearance levels) for each

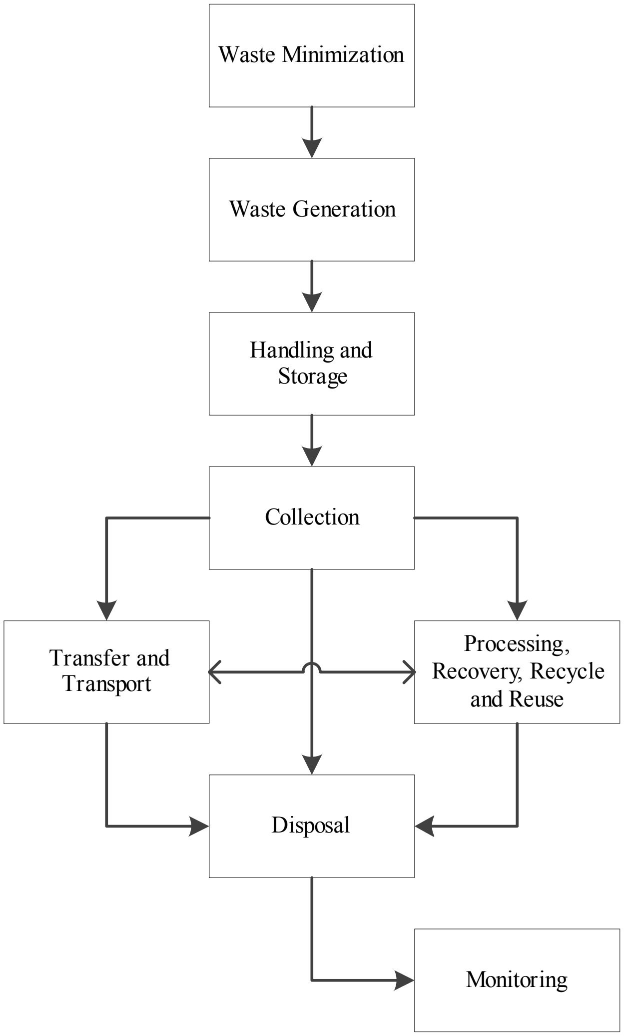

Fig. 2: Waste management operational functions.

radionuclide or decay series using GSS.

Generic clearance levels are very conservative. They are supposed to screen out wastes and industries from any further regulatory control and monitoring. Most industries like Iranian offshore petroleum industry are situated far from habitat areas, so both GSS and generic clearance levels would impose extra and unnecessary cost without any enhanced protection. In this case, specific clearance levels should be derived based on field observation and actual waste activity and management methods which occurs in reality.

Before developing Specific Scenario System (SSS) samples and data are gathered and their activity concentrations are measured. If their activity content is lower than generic clearance levels, the waste is cleared and could be released without any further regulatory control. Otherwise, wastes' fate must be investigated thoroughly with regard to exposure and environmental pathways and governing conditions of Lavan Island.

Formulation and justification of scenarios could be based upon relations and equation (Smith et al. 1996, IAEA 2003, IAEA 2005, EC 2001, Till & Grogan 2008) and any other appropriate reference. Before finalizing the results, an iterative approach might be required to calibrate the model. Calibration means that actual SSS does not satisfy dose criterion, so additional engineering designs like landfill barriers or protective measures like respiratory masks should be applied. These compartments are best determined by iterative approach which analyses the effect in SSS.

Integrated NORM management system would be eventuated by considering the assumptions and compartments which are applied to SSS in order to satisfy dose criterion and derive specific clearance levels. The final framework, which is applied to derive generic and specific scenario systems, is shown in Fig. 3.

RESULTS AND DISCUSSION

By evaluating these scenarios, generic clearance levels would be accomplished either for a country or a state. Tables 1 and 2 indicate the relation between the source term, exposure groups and waste management options in GSS and SSS respectively.

By comparing Tables 1 and 2, it is obvious that some source terms, waste management options and exposure groups are not valid for NORM management in Lavan Island. Following results could be concluded:

1. Exposure groups, waste management options and enclosed circumstances of Islands, make them a preferable location to distinguish between SSS and GSS and understand the approach of this paper.
2. Environmental pathways regard to food chains do not exist because no domestic animals live on the island. The only native terrestrial mammal is Chinkara which is not considered as food.
3. Landfilling of drums, which are shipped out of the island, does not fall in the investigation scope of specific assessment. Due to absence of landfilling option, no groundwater pathway and leaching parameters needs to be assessed. This would be a great advantage in preserving time and money, because soil sampling and hydrogeological studies require great amount of time and money.
4. Non-residential condition of Lavan Island would remove public exposure group from modelling and calculations. By changing the exposure group from public (usually considering children due to their high vulnerability) to workers (adults with age more than 17), the degree of

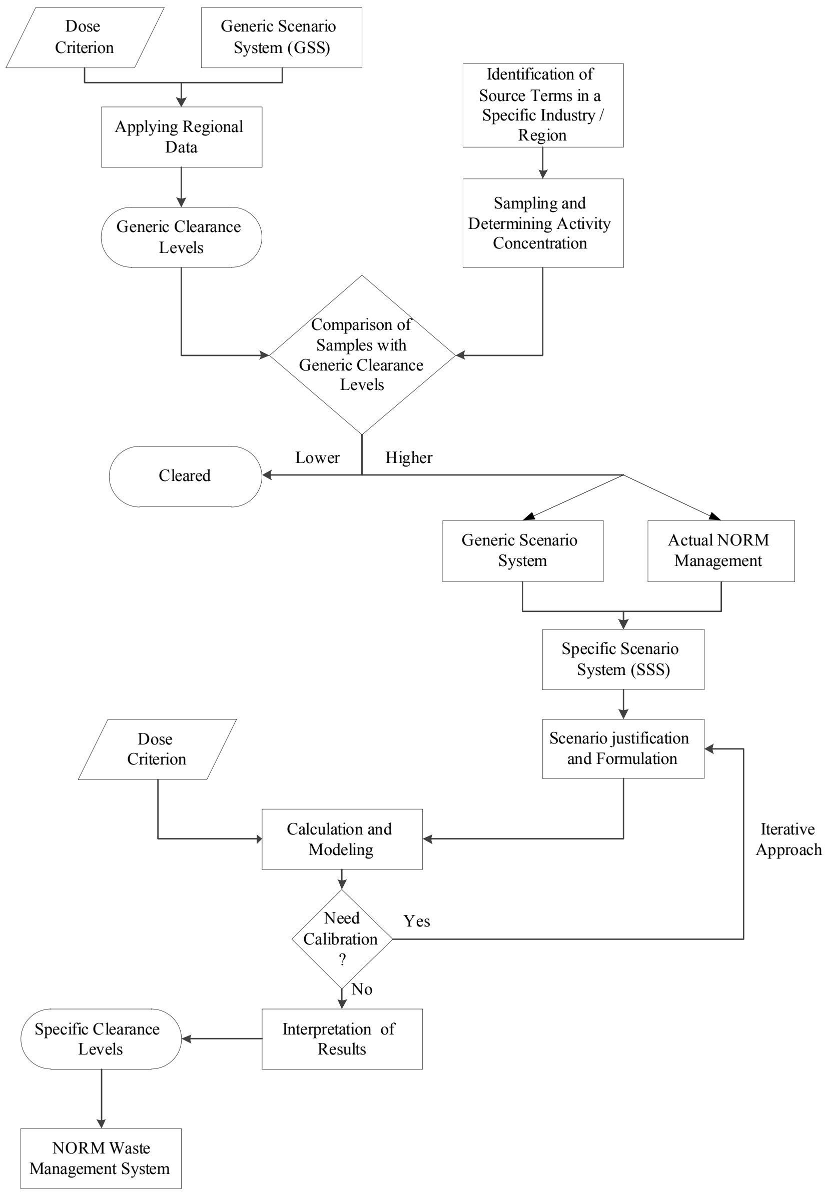

Fig. 3: Proposed new approach to derive generic and specific clearance levels.

pessimism and conservatism decreases. This leads to higher and more realistic clearance levels.

5. Generally, GSS is based upon conservatism assumption but SSS prospects real data. Specific clearance levels resulted from this approach maximizes health protection by minimizing the costs required to do so.
6. Life cycle assessment, which forms the main idea of this approach, completely covers all aspects of NORM management from cradle to grave.
7. It could be concluded that applying generic clearance levels for all industries (especially industries which are located far from habitat areas) is not recommended.

CONCLUSION

The authors have sought in this paper to highlight the importance of establishing both generic and specific clearance levels for NORM in each country due to different geological, hydrological, climate, food habits and cultures. The health protection requirements depend on internal policies and existence of proper infrastructures. It is not economically recommended for applying international regulatory context for all the countries and their industries especially the remote ones.

Fishing is one of the main activities on the island. Due

Table 1: Generic Scenario System (GSS).

		Waste Rock	Sand	Slag	Sludge	Scale	Ash	Liquid
Generation		W*	W	W	W	W	W	
Handling and storage		W	W	W	W	W	W	
Transportation		W	W	W	W	W	W	
3 R	House	W/P**	W/P	W/P				
	Playground	W/P	W/P	W/P				
	Road	W/P	W/P	W/P				
	Dam	W/P	W/P	W/P				
Controlled disposal	Landfilling	W/P	W/P	W/P	W/P	W/P	W/P	
Uncontrolled disposal	Open dumping	W/P	W/P	W/P	W/P	W/P	W/P	W/P
	Open burning				W/P			
	Discharging	P	P	P	P	P	P	P
Human intrusion		P	P	P	P	P	P	

Exposure group: *Worker / **Public

Table 2: Specific Scenario System (SSS) (Lavan Island).

		Waste Rock	Sand	Slag	Sludge	Scale	Ash	Liquid
Generation					W	W		
Handling and storage					W	W		
Transportation					W	W		
3 R	House							
	Playground							
	Road							
	Dam							
Controlled disposal	Landfilling							
Uncontrolled disposal	Open dumping				W	W		W
	Open burning				W			
	Discharging					W		W
Human intrusion								

to complexity and high degree of uncertainty, radiological assessment regarding to aquatic food chain should be carried out in an exclusive research.

In order to obtain better results and have an optimized framework, it is recommended to perform sensitivity and uncertainty analysis on exposure models. Adopting this policy needs complete and reliable data on local environmental parameters.

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