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DISPOSAL OF WASTE CONTAINING TECHNOLOGICALLY ENHANCED CONCENTRATIONS OF NATURALLY OCCURRING RADIOACTIVE MATERIALS – WHEN IS IT A CONCERN??

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ABSTRACT

Relatively consistent radioactive waste disposal criteria have been developed on an international basis [1]. This has resulted in a variety of different practices being accepted for disposal of short-lived radioactive wastes, primarily resulting from the nuclear fuel cycle. However, in the past 15 to 20 years increased attention has been drawn to a special category of radioactive wastes for which international agreement on regulation does not yet exist. This involves the large quantities of long-lived radioactive wastes contaminated with technologically enhanced concentrations of naturally occurring radioactive materials (TE-NORM). These wastes result from the extraction and processing of natural resources, such as oil and gas, coal and mineral resources as well as other activities. Different regulatory criteria are applied for TE-NORM in different countries and even in different States within a country (e.g., USA). Given the widespread occurrence of TE-NORM, it is desirable to have international consensus on appropriate technologies for disposal of these wastes.

This paper includes examples of TE-NORM wastes and different requirements which are being applied. Results of some simple calculations are provided to illustrate the potential hazards associated with disposal of TE-NORM and the dependence of those hazards on the method of disposal and the assumed habits of people living where the wastes are disposed of. External dose was chosen as the basis for the example calculations, because it is sufficient to provide an indication of the potential doses associated with disposal of TE-NORM waste while requiring minimal assumptions about environmental conditions and human habits.

INTRODUCTION

In the past 15 to 20 years, radioactive wastes from outside of the nuclear fuel cycle which are contaminated with technologically enhanced concentrations of naturally occurring radionuclides (TE-NORM) have developed from a little known issue to an issue that is receiving a large amount of global attention. This increase in the level of concern is reflected by the growing number of recent national and international conferences or symposia which have been focussed on TE-NORM (see for example [2,3]) and numerous publications addressing TE-NORM which have been prepared by regulators [4,5] and organizations such as the American Petroleum Institute (API) [6], United Kingdom Offshore Operators Association [7], European Commission (EC) [8,9], Gas Research Institute [10], Exploration & Production Forum (E&P Forum) [11] and the US National Academy of Sciences [12].

Two important reasons for the increasing levels of concern are: (1) the large amounts of TE-NORM wastes in many countries and (2) the potential long-term hazards resulting from the fact that TE-NORM is comprised of long-lived radionuclides with relatively high radiotoxicities. These two areas of concern have led to some difficulties in development of consistent standards for management of TE-NORM, which, in turn, has led to uncertainty in many countries regarding appropriate technologies for disposal of TE-NORM wastes. This paper provides some perspective regarding the potential doses associated with disposal of TE-NORM wastes on the ground surface.

Examples of sources and amounts of TE-NORM arisings and the radionuclides of concern in TE-NORM are introduced first in order to provide some perspective regarding the magnitude and scope of the problem. This is followed by some examples of standards and guidelines applicable to naturally occurring radionuclides. Some of these can be applied to TE-NORM wastes. Some example calculations are then provided to illustrate the potential doses to the public which can result from exposure to TE-NORM in cases with and without cover materials over the waste. This paper focusses on concerns for the general public and does not address the issue of protection of workers or non-human biota which may be exposed to TE-NORM. Furthermore, the document focusses on considerations relevant for current practices involving TE-NORM and does not address considerations in the context of intervention for contamination associated with disposal activities in the past.

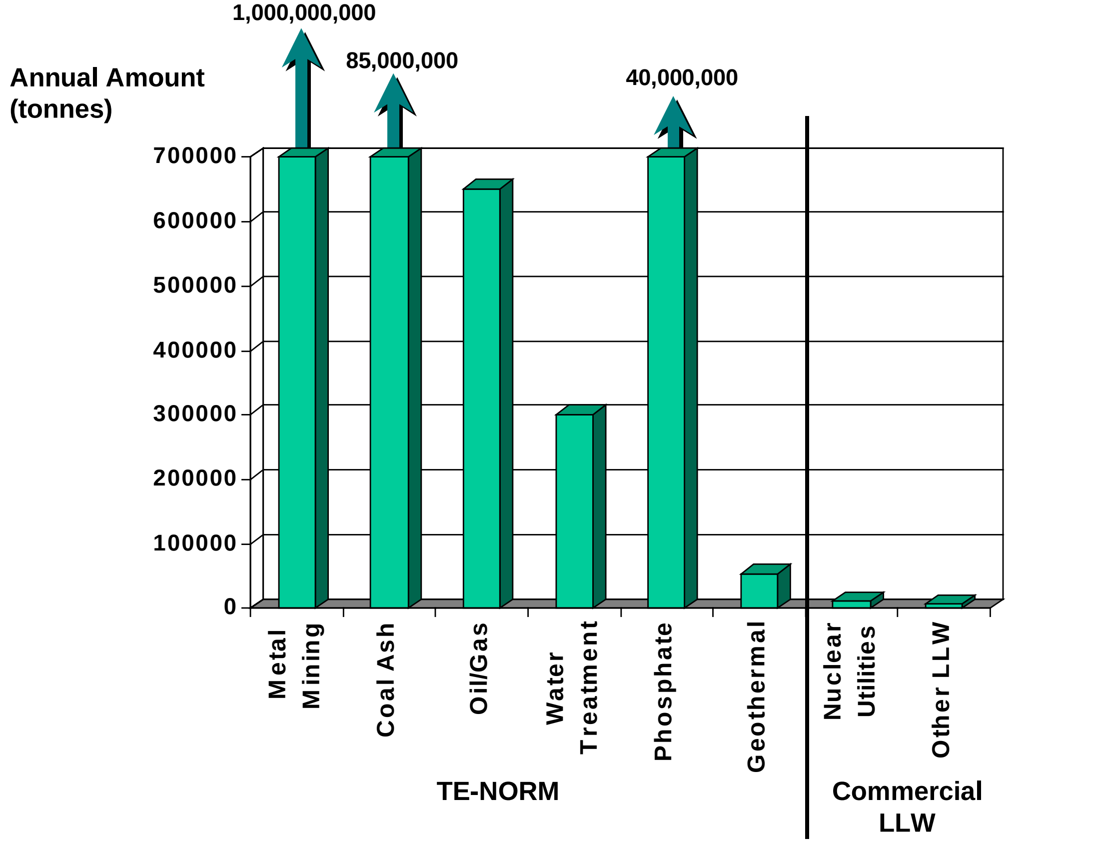
WHY ARE WASTES CONTAINING TE-NORM A POTENTIAL CONCERN FOR HUMAN HEALTH ?

TE-NORM comprises radionuclides associated with the U-238 and Th-232 decay chains as well as K-40, all of which have existed in the earth since its formation (see Table I). These radionuclides are very long lived and also have progeny which are long-lived, such as Ra-226. Radionuclides in these decay chains can also have a relatively high radiotoxicity. As shown in Table I, the ingestion dose factors of several radionuclides in these decay chains are relatively large. Furthermore, Ra-226 decays to Rn-222, a gas which has been recognized as a significant public health hazard, especially in cases where it can accumulate in homes. Given the long half-lives and the relatively large dose factors, TE-NORM poses potential health risks for long periods of time.

TABLE I. Naturally Occurring Radionuclides and selected data

Parent Radionuclide	Progeny	Half-Life (yr)	Ingestion Dose Factor (Sv/Bq) (e(g), adults)(1)
U-238+		4.5×10^{9}	4.8×10^{-8}
	U-234	2.4×10^{5}	4.9×10^{-8}
	Th-230+	7.7×10^{4}	2.1×10^{-7}
	Ra-226+	1.6×10^{3}	2.8×10^{-7}
	Pb-210+	2.2×10^{1}	1.9×10^{-6}
Th-232		1.4×10^{10}	2.3×10^{-7}
	Ra-228+	5.75×10^{0}	6.9×10^{-7}
	Th-228+	1.91×10^{0}	1.4×10^{-7}
K-40		1.3×10^{9}	6.2×10^{-9}

The US Environmental Protection Agency (US EPA) [4] has estimated that more than 1,000,000,000 tonnes of TE-NORM wastes are generated annually in the United States. The vast majority of the these wastes are the result of mining activities (see Figure 1). By comparison, the total amount of radioactive waste disposed of at commercial low-level waste disposal sites in the United States (USA) was on the order of ten thousand tonnes in 1997 [13], of which roughly 65% resulted from operation of nuclear power plants. As a point of comparison, based on estimates from the US EPA, the amount of TE-NORM resulting annually from oil and gas exploration and production activities in the USA may be on the order of several hundred thousand tonnes. The American Petroleum Institute has since produced a report [6], which suggests that the annual amount of TE-NORM from the oil and gas industry may be on the order of several tens of thousands of tonnes, still a substantial amount, but less than the estimates in the report for the US EPA. Although it is clear that there is a large amount of TE-NORM arisings, the key question is at what concentration of naturally occurring radionuclides do TE-NORM wastes become a concern for human health? The scope of the problem is critically linked to this consideration.



Note: Amounts of TE-NORM are rough estimates, amount of LLW is derived from reported disposal data at commercial disposal sites assuming 2 g/cm^3 of waste

Fig. 1. Estimated Annual Amounts of TE-NORM and Commercial LLW in the USA.

Table II includes examples of radionuclide concentrations in TE-NORM wastes from industries in Fig. 1. Concentrations on the order of 1 to 10 Bq/g are common in these wastes. These concentration levels can pose management challenges in terms of potential doses to the public. Given the fact that some of these industries can be found in most developing and industrialized countries, management of TE-NORM is a global issue rather than a problem of a small number of countries such as those which must deal with radioactive wastes from nuclear power production. This has led to concerns regarding management of these wastes in, for example, Southeast Asia and Africa, where countries are rich in natural resources and thus depend on industries associated with extraction and processing these natural resources (i.e., activities resulting in TE-NORM wastes).

TABLE II. Representative TE-NORM concentrations in selected materials [14]

Material	Radionuclide Concentrations (Bq/kg)
Scale in pipes and other equipment for handling oil/gas and formation waters	Background - 15,000,000 (average 1,000 to hundreds of thousand)
Sludges in natural gas supply equipment	Background - ~40,000
Sludges from ponds of produced water	10,000 - 40,000
Uranium mining overburden	100 - 20,000 (only Radium reported) (average of ~5,000 total radionuclide concentration)
Coal fired power plant ashes	200 - 25,000 (typically closer to lower value)
Drinking Water Treatment Waste	sludges - ~600 (only Ra-226 reported) resins - ~1,300,000 (only Ra-226 reported)
Phosphate fertilizer (biomass energy)	5,000 - 25,000
Other mineral processing waste (including aluminum, rare earths, etc.)	Background - 400,000 (generally 100 - 5,000)

Note: These values include maximums, averages for specific sets of data, or general ranges of values from a number of sources. In some cases, the radioactivity associated with only one or a few radionuclides was provided, when it is known that other radionuclides will be present. Thus, the table should only be used as a rough indicator. The data are compiled from a number of sources and summarized in [14].

INTERNATIONAL GUIDELINES AND REGULATORY STANDARDS

IAEA Basic Safety Standards

The International Atomic Energy Agency has published the International Basic Safety Standards for Protection against Ionizing Radiation and for the Safety of Radiation Sources (BSS) [1]. The BSS serve as a practical guide for development of regulations in individual countries. The BSS include a recommended limit of 1 mSv/yr for average doses received by members of a critical group as a result of practices involving radioactive materials. This limit applies to the sum of doses from all practices which may affect the critical group, thus, in most countries, the constraint applied to an individual practice is generally set at some fraction of 1 mSv/yr.

The BSS also include exemption levels on a radionuclide specific basis, which provide a basis upon which to determine if a radioactive substance can be exempted from the requirements specified in the BSS. On the surface, these values may appear to be a useful measure to determine when TE-NORM wastes should be subject to regulation as a radioactive waste. However, the BSS states that

"The application of exemption to natural radionuclides, where these are not excluded, is limited to the incorporation of naturally occurring radionuclides into consumer products or

their use as a radioactive source (e.g., Ra-226, Po-210) or for their elemental properties (e.g., thorium, uranium)."

Thus, the exemption values should not be applied in the case of disposal of TE-NORM as described in this paper.

This can be explained because of the basis for the exemption levels, which for the disposal case, were derived based on an assumption of placing a small source into a landfill [15]. The calculations involve large dilution factors and occupancy factors which may not be realistic for the case for disposal of TE-NORM. For example, the external dose calculations for the exemption values are based on an assumption that the radioactive materials are diluted in 10^{10} grams of non-radioactive waste and that a person is only exposed for 300 hours in a year. Furthermore, there is a probability of 0.01 for the external exposure scenario which is applied. In the case of TE-NORM, given the large amounts, it can be envisioned that persons could live on the materials (longer duration of exposure, probability of 1) and that the materials would be undiluted or only slightly diluted.

Regulations relevant for TE-NORM

Many individual countries and even individual States within countries have developed regulations for TE-NORM separate from general regulations for radioactive waste disposal. Requirements related to TE-NORM often take on a flavor of an exemption type value in individual countries rather than having TE-NORM subject to a site specific, dose based safety assessment for each practice [9]. For example, in Germany, NORM waste with a total specific activity less than 0.5 Bq/g are regarded as "non-radioactive". In the United Kingdom, separate values are provided for Actinium, Polonium, Protactinium, and Radium (0.37 Bq/g); Lead (0.74 Bq/g), Thorium (2.59 Bq/g) and Uranium (11.1 Bq/g). Concentrations of any isotopes above these values would result in the material being regarded as radioactive. The Netherlands regard TE-NORM with a total activity concentration greater than 100 Bq/g as radioactive (total activity limits also apply to limit the amount of material). In the United States requirements for TE-NORM are being specified for the most part by the individual States. In general, the targets for release from control have been set at total activity concentrations less than roughly 1.1 Bq/g (30 pCi/g) or 0.2 Bq/g (5 pCi/g).

The Netherlands requirements are based on Directive 84/467/Euratom which was promulgated by the Commission of European Communities (CEC) in 1984. The CEC promulgated a new radiation protection directive (96/29/Euratom) in 1996 which essentially mirrors the IAEA BSS. However, it should be noted that exemption values in the CEC directive are based on the same calculations as those used for the IAEA BSS, thus, they should not be applied for TE-NORM. The primary concern is that many countries have not developed requirements related to disposal of TE-NORM. The lack of consistency in requirements being applied for TE-NORM is a source of confusion for countries trying to develop requirements of their own.

WHAT ARE THE POTENTIAL HEALTH HAZARDS ASSOCIATED WITH DISPOSAL OF TE-NORM?

Given that there is some disparity in the requirements, it is informative to consider some calculations to illustrate the potential hazards to human health associated with disposal of wastes containing TE-NORM. Some calculations were recently conducted for the US

Department of Energy in order to assess the potential health effects associated with landspreading (e.g., mixing in surface soil) of TE-NORM from the oil and gas industry [16]. The results of the calculations suggested that practices resulting in concentrations of Ra-226 in surface soils greater than 0.37 Bq/g (10 pCi/g) should be evaluated on a case by case basis to estimate potential future risk. This conclusion was based on the results of calculations which suggested that critical group exposures to concentrations of Ra-226 greater than 0.37 Bq/g can result in doses in excess of 1 mSv/year for a residential scenario that could reasonably be expected to occur. Furthermore, the results suggested that landspreading of TE-NORM which yields concentrations of Ra-226 greater than 0.37 Bq/g (10 pCi/g) may involve the need to include restrictions on future use of areas where such landspreading has occurred. Placement of a "clean" cover over the TE-NORM waste was shown to provide some benefit at early times, but given the long half-lives of radionuclides in TE-NORM, erosion would be expected to remove a basic cover before the radionuclides have decayed significantly.

The calculations discussed above were conducted for living conditions expected in the United States. When identifying appropriate management approaches, it is important to reflect conditions expected in the region where the waste would be disposed. In tropical regions, especially in rural areas, it is common for people to live in homes which allow a large amount of air circulation and may involve the use of soil or bricks made from local soil as construction materials. These are important distinctions, because increased air circulation reduces the possibility for Rn-222 to build up in a home (i.e., doses due to Radon may be reduced). However, shielding assumptions will be different for external dose calculations, especially if local soil which may include TE-NORM is used as a construction material (i.e., less shielding would be expected and external doses may be enhanced if TE-NORM is in the construction materials). Another factor which will be explored is the dependence of the dose on the amount of time that people spend in and around their home. In some rural areas, it is common for people to spend more time near their home, especially outside the house.

Example calculations

A few simple calculations were conducted to illustrate the potential doses associated with disposal of TE-NORM and the sensitivity of the predicted doses to assumptions about living conditions and the disposal practices. The intent was to illustrate the potential hazards to human health associated with disposal of TE-NORM as well as factors which help determine the magnitude of those hazards. To minimize the input requirements and thus debate about values selected for inputs, the calculations focus on the external dose pathway. The external dose pathway provides a sufficient illustration for the purposes of this paper and also involves the least amount of assumptions regarding the environmental conditions at the site and the habits of the critical group. A complete dose assessment would need to consider all pathways deemed appropriate (e.g., groundwater consumption and use, dust inhalation, ingestion of contaminated food products (vegetables, meat, milk), etc.). As shown in Table I, some natural radionuclides have high ingestion dose factors and thus, a formal assessment would also need to consider this pathway. Furthermore, radon has been identified as a significant health hazard, so it may be necessary to address doses for radon inhalation as well.

The RESRAD computer code [17] was used for the calculations. RESRAD has been used extensively in the United States and internationally for assessments of doses associated with radionuclides in soils. Although RESRAD has the capability to consider a number of different pathways for exposure, as discussed above, all pathways except external dose were disabled for the calculations presented here. As mentioned above, this pathway is sufficient to illustrate the potential hazards, while only involving basic assumptions regarding environmental conditions and human habits. The equation used to assess the external dose at a given time is

$$Dose_i = C_i \times Occupancy \times Df_{ext,i} \quad (1)$$

where:

$Dose_i$ is the dose to a member of the critical group from radionuclide *i* (mSv/yr)
C_i is the concentration of radionuclide *i* in the contaminated soil (Bq/g)
Occupancy is the occupancy factor (dimensionless)
$DF_{ext,i}$ is the external dose factor for radionuclide *i* (mSv/Bq/g).

The occupancy factor is defined as

$$Occupancy = indoor \times shield + outdoor \quad (2)$$

where:

indoor is the fraction of the time spent inside the home in the contaminated area
shield is the shielding factor which represents the fraction of the external radiation which is assumed to penetrate the walls and floor of the home
outdoor is the fraction of time spent outside the home in the contaminated area.

The input data used for the calculations are summarized in Table III. All of the calculations are based on an initial concentration of 1 Bq/g of Ra-226 and 1 Bq/g of Th-232 in surface soil to a depth of 0.15 m in an area of 1000 m^2. This represents roughly 150 m^3 of contaminated soil, which is actually a small amount of TE-NORM (a fraction of 1000 tonnes, undiluted). The use of a unit concentration facilitates easy conversion of the results to a specific concentration for a specific waste. All progeny are assumed to reach equilibrium with the parent as determined by decay.

TABLE III. Summary of different cases considered.

	Case 1	Case 2	Case 3
Radionuclide	Ra-226+, Th-232+	Ra-226+	Ra-226+
Cover thickness	none	0.3, 0.5 and 1 m	none
Occupancy factor	0.6	0.6	0.3, 0.6, 0.9

Case 1 is a base case to illustrate potential doses for both Ra-226+ and Th-232+ for reference conditions of no cover and an occupancy factor of 0.6. From Equation 2, an occupancy factor of 0.6 could represent a person that spends 50% of their time inside a home in which 70% of the radiation penetrates the walls and an additional 25% of their time outside the home but still in the contaminated area. Case 2 focusses on sensitivity of the results for Ra-226 plus progeny to the depth of cover over the waste assuming an occupancy factor of 0.6. The cover is assumed to be made of clean soil with a density of 1.5 g/cm^3. Case 3 addresses the sensitivity of the results to the value of the occupancy factor. The low value for the occupancy factor would represent the case of a critical group that is frequently away from home and (or) spending time in a home that provides substantial shielding and also minimal time outside the home in the disposal area. The large value for the occupancy factor could represent a rural situation where the critical group lives in a home providing minimal shielding and also works in the contaminated area (e.g., rural farmer). Note that the occupancy factor is a linear multiplier in Equation 1.

Results

The results of the simulations are presented graphically in Figures 2 through 4. For the reference conditions, the results in Figure 2 suggest that an initial concentration of 1 Bq/g of Th-232 or Ra-226 in TE-NORM wastes placed on the ground surface can result in external doses in excess of 1 mSv/year (assuming progeny grow in over time). Figures 3 and 4 illustrate the results of Cases 2 and 3, respectively. Figure 3 illustrates the effectiveness of a cover in reducing the external dose at early times (only Ra-226 + progeny). However, when the cover erodes, which in many cases occurs before significant decay of the radionuclides, the doses exceed or approach 1 mSv/year. This highlights one of the fundamental problems with near surface disposal of long-lived radionuclides at concentrations which can pose significant health effects. Namely, the radionuclides will generally outlast any basic barriers.

Figure 4 illustrates the effect of assumptions regarding human habits near the waste (Ra-226 + progeny for uncovered waste). The only case which results in doses consistently below 1 mSv/yr is for a very small occupancy factor of 0.3. This reflects conditions where the resident would spend minimal time near or in their home assuming that the home provides some shielding. These results illustrate that for uncovered waste, most occupancy assumptions can easily result in doses above 1 mSv/year.

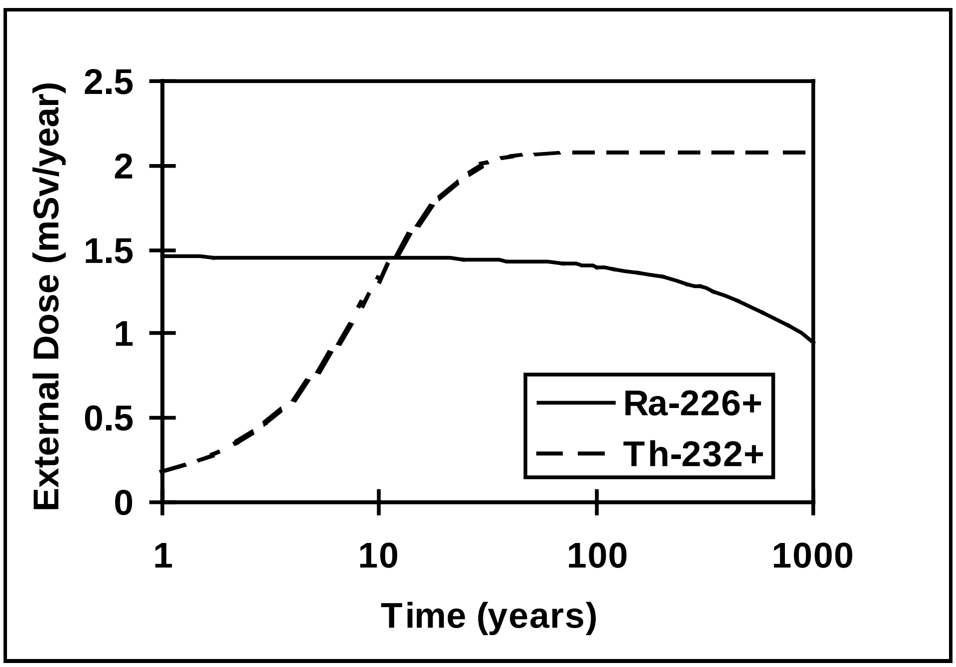


Fig. 2. Results for Case 1 (Note that the external dose for Ra-226+ is dominated by Bi-214 and the external dose for Th-232+ is dominated by contributions from Tl-208 and Ac-228).

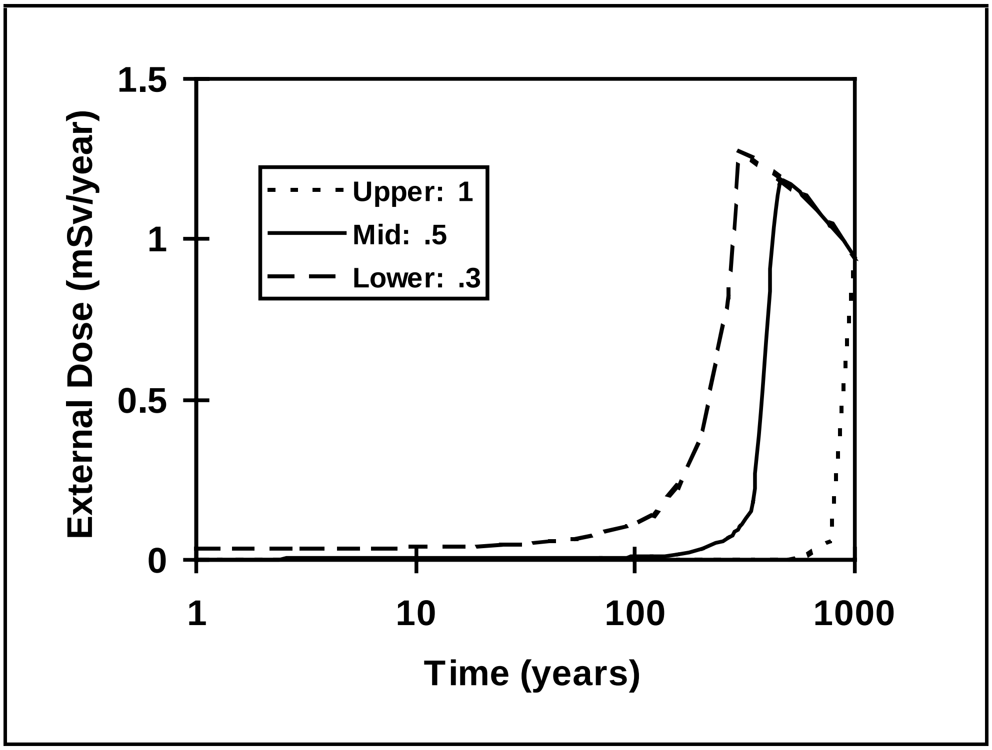


Fig. 3. Results for Ra-226+ for different amounts of cover (m) over the TE-NORM.

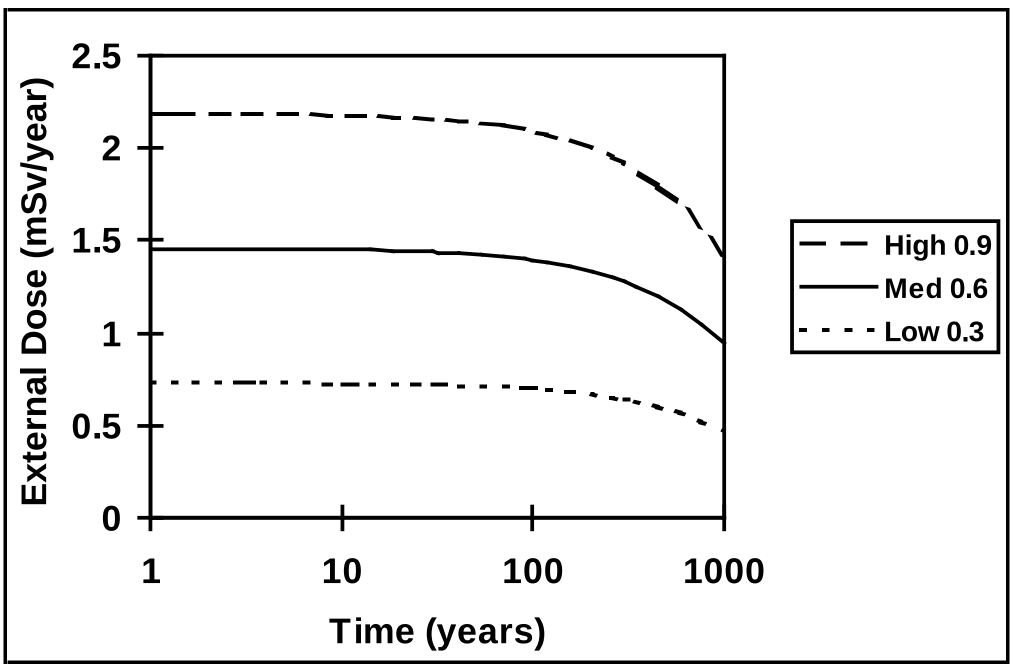


Fig. 4. Results for Ra-226+ for different occupancy factors.

The results of the example calculations illustrate that initial concentrations of 1 Bq/g of Ra-226 or Th-232 in a relatively small amount of TE-NORM wastes disposed on the ground surface can result in doses in excess of the limits identified in the IAEA Basic Safety Standards (1 mSv/year) even when only considering external exposure. Many TE-NORM wastes comprise concentrations of radionuclides greater than 1 Bq/g (see Table II). Placement of a cover over the waste can help delay the effects, but as erosion occurs, the wastes may eventually be exposed and lead to doses in excess of regulatory limits. Given the amount of TE-NORM wastes which are generated annually, robust disposal facilities are often not an economically viable option because of the impact on key industries.

CONCLUSIONS

TE-NORM wastes are generated in numerous countries, especially those that are rich in natural resources such as oil, natural gas, coal or metals/minerals. The amounts of these wastes are very large relative to the amounts of radioactive waste from other sources. Furthermore, TE-NORM involves radionuclides which can pose relatively high hazards for long time frames. In the past 15 to 20 years the attention on the potential hazards associated with these wastes has grown to the point where they are the subject of concern on a global level. Different countries and even different states within a country (e.g., the USA) can have different activity concentration levels below which TE-NORM is not considered a significant radiological health hazard. In many countries, regulations for control of activities involving TE-NORM are not in place or are only in the process of being prepared.

Some example calculations were provided to illustrate that given the radiotoxicity and relatively long half-lives associated with radionuclides in TE-NORM, relatively low activity concentrations (1 Bq/g) can result in doses in excess of 1 mSv/yr to a member of the general public, even when only the external exposure pathway is considered. The doses can be larger when other pathways are considered (e.g., radon inhalation, drinking water). Placing a clean cover (0.3 to 1 m) over the TE-NORM waste was shown to help reduce the doses over early times, but as the cover erodes, the long-lived radionuclides can still pose a hazard a thousand years or more into the future. Short of mandating land use restrictions for potentially thousands of years, these doses pose a problem in identifying appropriate disposal technologies for the large amounts of TE-NORM wastes that are produced each year. Economic considerations when selecting appropriate disposal technologies are an important factor for these wastes.

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