Interim Guidelines for Disposal of Solid Waste containing TENORM and Arsenic resulting from Drinking Water Treatment Processes

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1.1. Definitions (Adapted from Health Canada, 2000):

(a) Becquerel (Bq) is a unit used for measuring the quantity of radioactivity present without consideration to the type of radiation emitted. (1Bq = 1 nuclear transformation/sec, 1Bq = 27 pCi [pico-curie]); 1nCi = 1000 pCi

(b) Occupationally exposed workers are employees who are exposed to NORM sources of radiation as a result of their regular duties;

(c) Sievert (Sv) is a unit of Effective Dose of radiation and accounts for the total effect of different types of radiations on different parts of the body. Regulations express the dose on a yearly basis, as millisieverts per annum (mSv/a);

(d) Incidentally exposed worker are employees whose regular duties do not include exposure to NORM sources of radiation. They are considered members of the public who work in an occupational exposure environment.

1.2. Introduction

Water Treatment Plant (WTP) operators derive their raw water supply from surface water bodies such as rivers, streams and reservoirs, or from underground sources. Federal (Health Canada, 2008) and Provincial Standards (EPB 507) require water suppliers to limit the occurrence of contaminants such as metals, pesticides, toxic substances including radionuclides in drinking water. Various processes are employed to treat raw water in order to comply with these regulations e.g. coagulation/ filtration, precipitative softening, membrane separation, ion exchange, granular activated carbon and stripping process. During the process of water treatment, these contaminants get accumulated in the waste stream in significant concentrations that require special handling. The type and the quantity of solid waste generated primarily depend on the treatment process used, the raw water quality and its source. The presence of radionuclides in WTP residual waste does not make it hazardous; hazardous waste generation will most likely be the result of the removal of certain co-occurring contaminants such as arsenic, in the residual waste (USEPA, 2005).

This document is intended to provide guidelines for proper disposal of solid residuals resulting from drinking water treatment processes that may be contaminated with arsenic and/or radioactive material, the latter being termed as Technologically Enhanced Naturally Occurring Radioactive Material (TENORM). Such residual waste typically comprises spent resins, spent filters media, spent membranes and sludge.

1.3. Sources of Contaminants in the Environment

1.3.1. Arsenic

Arsenic occurs in the environment both from natural and anthropogenic sources. The primary natural sources of arsenic releases to the environment are: hot springs (geothermal), igneous rock (basalt), sedimentary rock (organic/inorganic clays, shale), metamorphic rock (slate), seawater, mineral deposits; and volcanoclastic materials/releases (USEPA, 2003). Major anthropogenic source for the release of arsenic to the environment includes pesticides, solid waste disposal, mine tailings, industrial emissions, wood preservatives and combustion byproducts from burning fossil fuels (predominantly coal). Arsenic exists in the environment both, as inorganic and organic species. Inorganic arsenic can occur in the environment in several forms but in natural waters and so in potable water, it mostly exists as arsenite, As(III) and arsenate, As(V) (USEPA, 2000). Organic arsenic species are although abundant in seafood but they are relatively much less harmful to health than inorganic arsenic and are readily eliminated by the body. The forms of arsenic present will depend on the type and amounts of sorbents, pH, redox potential (Eh), and microbial activity in the environment where it exists (Yong & Mulligan, 2006).

Arsenic is a documented human carcinogen and has been classified in Group 1 of the Priority Substance List which identifies substances including chemicals, effluents and wastes that are harmful to the environment or constitute a danger to human health (Health Canada, 2006). Across Canada the arsenic concentrations in natural uncontaminated soil and sediments range from 4 to 150 mg/kg whereas in uncontaminated surface water and groundwater, the concentration ranges from 0.001 to 0.005 mg/L (Wang & Mulligan, 2006). However, much elevated levels, ten to thousand times the Interim Maximum Acceptable Concentration (IMAC) as a result of anthropogenic inputs, have been reported in air; soil and sediments; surface water and groundwater; and biota in several regions of Canada depending upon local geology, hydrogeology and geochemical characteristics of the aquifer (Wang & Mulligan, 2006).The regulatory limit for Arsenic in drinking water as of December 2010 is 0.010 mg/L.

1.3.2. NORM [TENORM]

Naturally Occurring Radioactive Materials (NORM) is present in trace concentrations in surface waters, groundwater, soils and rock and comes into existence as a consequence of radioactive decay of uranium (U-238) and thorium (Th-232). Uranium is mobilized from rock by the weathering of uraninite (UO_2) and by the action of surface waters and groundwater oxidative dissolution of uraninite into uranyl ion (UO_2^{2+}) (Canada Gazette, 1998). Major anthropogenic activities that release uranium into subsurface environment are uranium mining and milling, uranium processing, phosphate mining, heavy

metal mining, coal use and inappropriate waste disposal. Other common radionuclide of concern that may be found in groundwater includes radon-222, radium-226, and radium-228 which are the byproducts of U-238 and Th-232 decay series.

Most TENORM is produced incidental to an industry's main products as a result of human activities. These industrial processes lead to alteration or enhancement of physical, chemical, radiological properties and concentrations of NORM such that a potential exists for (CRCPD):

(i) Redistribution and contamination of environmental media (soil, water and air);

(ii) Incorporation of elevated levels of radioactivity in products and construction material;

(iii) Relatively high exposures to individuals and populations via any environmental pathway and medium due to its improper disposal.

In general, TENORM is recognized as being of two types:

(i) Discrete TENORM that has a relatively high radioactivity concentration/activity distributed in a very small volume (e.g. radium source used in medical procedures).

(ii) Diffuse TENORM that has a much lower concentration of radioactivity distributed over a large volume of material (e.g. contaminated soils and WTP residuals).

Drinking water supplies in Canada usually have uranium concentrations less than 0.01 mg/L, but values as high as 0.70 mg/L in private domestic supply in Nova Scotia have also been reported (Canada Gazette, 1998). A wide range of radionuclide concentrations may be observed across Saskatchewan depending on the water source; however, the drinking water standards for Saskatchewan (EPB 507, 2012) limit maximum acceptable concentration (MAC) of uranium to 0.02 mg/L, gross α-activity to 0.5 Bq/L and gross β-activity to 1.0 Bq/L in drinking waters.

1.4. Mobility of Contaminants in Landfill Environment

1.4.1. Arsenic

Mobility of arsenic in landfills is affected not only by initial leaching, but also influenced by adsorption; reactions mediated by redox and pH conditions; and biological activity. Microorganisms can affect the distribution of arsenic by accumulating, transporting, and transforming it. Under anaerobic conditions, arsenic compounds are reduced and methylated to di- and tri-methylarsine by various anaerobes (Jing *et al.,* 2005; Ferguson & Gavis, 1972). Adsorption is a pH dependent phenomenon and in case of arsenic, adsorption will be least at lower pH values for As (III) and greatest at neutral pH for As (V) (Pinel-Raffaitin *et al*., 2007; Blakey, 1984).

The Toxicity Characteristics Leaching Procedure (TCLP –method 1311, USEPA), which is conducted to identify whether the hazardous components of waste are likely to leach out and become a threat to public health or the environment (USEPA, 2006), does not adequately simulate the alkaline pH, low redox potential, biological activity, long retention time and organic composition typical of landfill environment (Ghosh *et al.*, 2004). All these environmental parameters favor mobilization of arsenic from metal oxide sorbents. As a matter of fact, based on TCLP, most arsenic bearing solid residuals from water treatment processes may be disposed in a non-hazardous landfill (Ghosh *et al*., 2006). It is therefore important to recognize this fact and recommended to conduct leaching test for arsenic under conditions that are typical of landfill environment prior to its disposal in a non-hazardous landfill.

1.4.2. NORM [TENORM]

The potential for surface water and groundwater contamination as well as direct exposure to NORM will depend on the method chosen for its disposal and the environment where it is disposed (IAEA, 2003). Figure 1 shows various potential pathways for the transport of TENORM. When the disposal is planned at landfill, the leaching and migration of radionuclides is influenced by subsurface hydrogeology and the chemical properties of the medium (soil and groundwater) with which it interacts (Pulhani *et al*., 2007).

Solubility of uranium and others is very site specific and has been reported to be the net result of competition between complexation with carbonates, hydroxyls, phosphates, sulphates; and precipitation and sorption on solid phases (De Windt *et al*., 2003). Uranium is strongly adsorbed onto the iron oxide and therefore soils enriched with iron oxide effectively immobilize uranium before it can reach the underlying aquifers (Brown *et al.*, 1998). Similarly Radium-226 other than dispersed or dissolved, is adsorbed and retained in the soil and does not migrate through the soil (Boscov *et al*., 2001). However the presence of carbonates and sulphates in soil increases the mobility of uranium (Pulhani *et al*., 2007).

Although the possibility of leaching of radionuclides from disposal of WTP residuals in the landfill environment does exist, routine surveillance of groundwater from disposal sites has shown that the levels of these radionuclides do not exceed the recommended WHO limits (Pulhani *et al*., 2007). Based on above discussion, it may be evident that mobility of TENORM is not a major concern when the WTP residuals are disposed in landfills; however, its radioactivity should decide its disposal option.

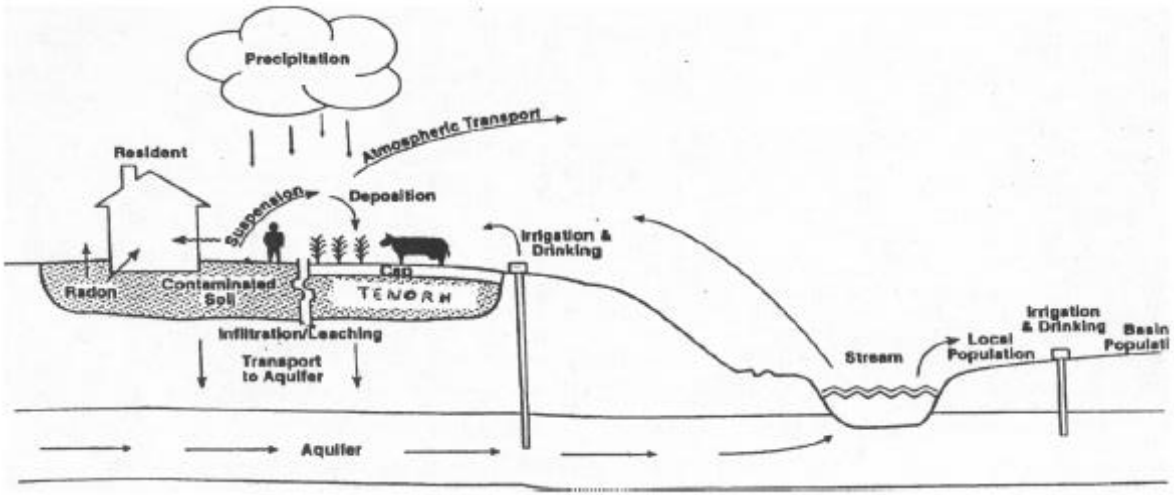


Figure 1 Environmental Transport Pathways for TENORM (Source: CRCPD)

1.5. Statutes & Regulations

NORM does not come under the control of Canadian Nuclear Safety Commission (CNSC) except for its import, export and transport. The transportation of NORM with radioactivity below 70Bq/g is not subject to federal transportation regulations (OH&S, Ministry of Labour). Above 70 Bq/g, the radioactive material is subject to the requirements of federal regulations including CNSC Packaging and Transport Regulations (Canada Gazette, 1998); Transport of Dangerous Goods Regulations (Canada Gazette, 1980) and IAEA Regulations for Safe Transport of Radioactive Materials (IAEA, 2003). Provincial guidelines (Energy & Resources, 2003) and regulation (Environment, 2000) do provide for handling/ management of NORM and hazardous substances respectively, however, regulation for disposal of TENORM resulting from drinking water treatment processes is not specifically spelled out both in Federal and Provincial regulation. Until such time specific provincial regulations for management of TENORM are enforced, it is recommended that the guidelines presented herein shall be followed.

1.5.1. Background Radiation & Recommended Radiation Dose Limits

In Canada on an average, a person may receive a range of annual doses from background radiations from 1.2 mSv to 3.2 mSv (Health Canada, 2000) which comes from:

(i) cosmic radiations from sun and outer space,

(ii) environmental radiation coming from natural radioactivity at the earth's surface: and

(iii) internal radiation e.g. from inhalation of radon progeny and ingestion of radiation from muscle tissues containing potassium.

In addition to background radiation dose, an annual NORM radiation dose limit of 20 mSv for occupationally exposed workers; and 1 mSv for incidentally exposed workers and members of the public has been recommended (Health Canada, 2000). To further ensure that the public and incidentally exposed workers do not exceed the annual dose limit of 1 mSv, an annual dose constraint of 0.3 mSv has been adopted for unconditional release into the public domain (Health Canada, 2000). Since landfills may not be considered as a public domain, a maximum annual dose limit of 1.0 mSv that will provide safety for incidentally exposed workers and members of the public has been considered in this guideline for calculating the radioactivity of the residual waste in determining its disposal option.

1.6. Types & Concentration of Residual Waste

As discussed in foregoing paragraphs, the type and quantity of radionuclide in solid residuals also depend on the ability of the WTP to remove specific radionuclides. Table 1 lists various drinking water treatment processes, the radioactive contaminant that they remove, and the types of residuals. Radon is a gas than can be removed from drinking water by air stripping and GAC, neither of which produces a residual for routine disposal. Furthermore, radon has a very short half-life of approximately 3.8 days and decays to essentially zero in roughly 38 days. Therefore, radon should not be found in any waste stream from a WTP except in the air from an air stripper (USEPA, 1996).

Information about typical concentration of arsenic in WTP residuals has not been well documented and is not available at the time of preparation of this document. A limited amount of information has been reported on the concentration of radionuclides in the waste streams of several water treatment processes. Table 2 presents radionuclide concentration observed in residual waste from few WTPs across North America (USEPA, 1996). The data presented in Table 2 are site-specific and are only intended to provide an approximate range of radionuclide concentration that may be expected in these waste streams. Actual measurement of concentration, however, should always be undertaken before deciding its disposal option.

Table 1: Water Treatment Process materials containing Radionuclide (Source: USEPA, 1996)

Treatment Process	Radionuclide removed	Process Material
Coagulation / Filtration	Radium, Uranium	Filter medium (sand) Filter medium (coal)
Lime Softening	Radium, Uranium	Filter medium (sand) Filter medium (coal)
Cation exchange	Radium	Resin
Anion exchange	Uranium	Resin
Iron removal processes: (a) Oxidation / Filtration (b) Greensand adsorption	Radium	Filter medium (sand) Filter medium (coal) Greensand
Reverse osmosis	Radium, Uranium	Membrane
Electrodialysis	Radium, Uranium	Membrane
GAC adsorption	Radium, Uranium, Radon	GAC
Selective sorbents	Radium, Uranium	Selective sorbent media

Table 2: Concentration of radionuclides on water treatment process media and materials (Bennett, 1978; Brink et al. 1978): Source USEPA (1996)

Location	Treatment Process	Process Media/Material	Radionuclide Ra-226	Concentration
Lamont, IL (Pilot Study)	Radium selective complexer	Resin	Ra-226 Ra-226	3.6 nCi/g (exhaustion) (total radium)
Redhill Forest, CO	Radium selective complexer (brine treatment)	Resin	Ra-226	$770x18^8$ pCi/ft^3
Herscher, IL	Iron removal	Filter media	Ra-226 Ra-228	111.6 pCi/g 38.9 pCi/g
	Cation exchange	Resin	Ra-226	43 pCi/g
Lynwood, IL	Cation exchange	Resin	Ra-226 Ra-228	9.6 pCi/g 6.6 pCi/g
Dwight Correctional Institute, IL	Natural greensand (cation exchange)	Greensand	Ra-226	29–46 pCi/g
Peru, IL	Lime softening	Filter media	Ra-226 Ra-228	4.6 pCi/g 3.6 pCi/g
Elgin, IL	Lime softening	Filter media	Ra-226 Ra-228	16.0 pCi/g 8.3 pCi/g
Elkhorn, WI	Iron removal	Filter media (sand)	Ra-226 Ra-228	1.47 pCi/g 0.48 pCi/g
	Cation exchange	Resin	Ra-226 Ra-228	6.04 pCi/g 2.7 pCi/g
Mt. Vernon, NH	GAC Radon removal	GAC	U-235/238 Pb 210	549–9,050 pCi/g (top of bed) 757 pCi/g (max)
Amherst, NH	GAC Radon removal	GAC	Pb-210	297 pCi/g (max) —

Key
GAC = granular activated carbon.

1.7. Residual Waste Characterization

Characterization of waste is an important step prior to determining its disposal options. The residual waste from water treatment processes should be characterized for its radioactivity (Bq/kg) and co-contaminants as per Table 3 using TCLP. The owner of the water treatment plant must ensure that the residual waste is tested, and disposed in accordance with these guidelines.

Table 3: Maximum concentration of contaminants for the Toxicity Characteristics (USEPA, 2006)

Contaminant	Concentration (mg/L)	Contaminant	Concentration (mg/L)
Arsenic	5.0	Chromium	5.0
barium	100.0	Lead	5.0
Cadmium	1.0	Mercury	0.2
		Silver	5.0

1.8. Disposal Options

Given the long-half-lives associated with NORM, institutional control at disposal facilities may need to be maintained for a significant length of time. As discussed above and on the basis of annual maximum dose limit (1.0 mSv/a); unrestricted disposal of TENORM containing waste may be allowed at any landfill provided annual radioactivity measured in the residual waste does not exceed the values shown

in Table 4. The values shown in Table 4 that will deliver a maximum effective dose of 1.0 mSv/a under conservative scenarios were obtained using a linear extrapolation of values provided by the Federal Provincial Territorial Radiation Protection Committee for dose constraints of 0.3 mSv/a (Health Canada, 2000). The limits indicated in Table 4 for U-238, Th-230, Th-232 & K-40 have been rounded off as per telephonic communication with the Ministry of Labour.

Where more than one long-lived radionuclide is present in a sample, the appropriate sum of the ratios of the radioactivity of each long-lived radionuclide and its corresponding Release limit as per Table 4 must not exceed 1 as shown below (Health Canada, 2000).

$$\left[\frac{\text{NORM- A concentration}}{\text{Unconditional derived release limit for NORM A*}} + \frac{\text{NORM- B concentration}}{\text{Unconditional derived release limit for NORM B*}} + \cdots\cdots + \frac{\text{NORM- n concentration}}{\text{Unconditional derived release limit for NORM n*}}\right] \leq 1$$

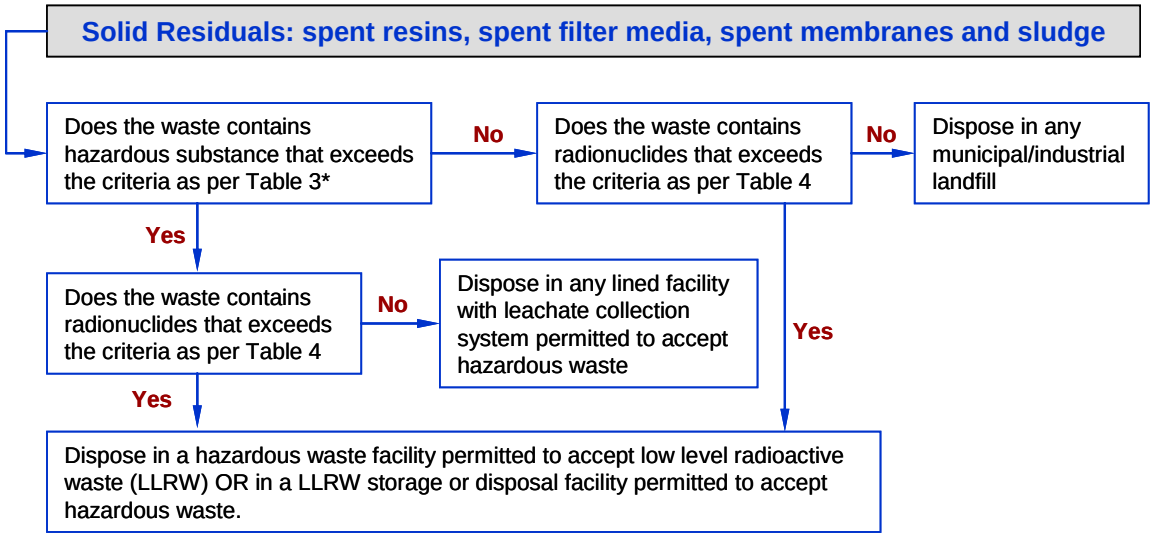
* Limits as per Table 4

Table 4: Annual Unconditional Derived Release Limits for Landfills – Diffuse NORM Source (Modified from Health Canada, 2000)

NORM Radionuclide	Derived Release Limit for solid residual waste that will provide a maximum dose limit of 1.0 mSv/annum (Bq/kg)
Uranium-238 series (all progeny)	1000
Uranium-238 (U-238, Th-234, Pa-234m, U-234)	30000
Thorium-230	30000
Radium-226 (in equilibrium with its progeny)	1000
Lead-210 (in equilibrium with bismuth-210 and polonium-210)	1000
Thorium-232 series (all progeny)	1000
Thorium-232	30000
Radium - 228 (in equilibrium with AC-228	1000
Thorium-228 (in equilibrium with all its progeny)	1000
Potassium-40	50000

In situations where the radioactivity in the residual waste exceeds the limiting value shown in Table 4, the disposal options shall be decided as per Figure 2 .

The low-level radioactive waste (LLRW) facility as mentioned in Figure 2 is regulated by the CNSC who makes the licensee responsible for the safety of the facility and establishes a number of guidelines to which the operator of the facility must adhere. In general, it is the responsibility of the licensee to explain to the CNSC how it plans to meet its performance criteria, and how safety will be assured. Considerable effort will be needed to ensure that the public has input into decisions, both on the type of waste facility needed and where it should be located.



* In addition to EPA Method 1311, leaching of contaminants should also be conducted under conditions typical of landfill environment.

Figure 2: Flow Chart for determining disposal option for solid residuals resulting from drinking water treatment processes

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