

Incidental TENORM: A Guidance for State Solid Waste Managers

FINAL

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Radiation Focus Group Federal Facilities Research Center

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ASTSWMO's mission is to enhance and promote effective State and Territorial programs for waste and materials management, to encourage environmentally sustainable practices and to affect national waste and materials management policies.

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The Association of State and Territorial Solid Waste Management Officials (ASTSWMO) is an organization supporting the environmental agencies of the States and Territories (States). ASTSWMO's mission is to enhance and promote effective State and Territorial programs for waste and materials management, to encourage environmentally sustainable practices and to affect relevant national waste and materials management policies. This document was prepared by the ASTSWMO Federal Facilities Research Center's Radiation Focus Group. The mission of the Focus Group is to identify national level radiation issues, coordinate State input, encourage improved partnership between State and Federal Agencies; and produce issue papers and other products as necessary to promote State interests on national radiation issues involving site cleanup and health and safety at federal facilities and other sites. The group acts as a resource to States in researching issues regarding radiation, providing information to States, and assisting in building State radiation program capacities, as requested by members.

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1.0 SCOPE

The purpose of this guidance is to inform solid waste management and other officials about technologically-enhanced, naturally-occurring radioactive materials (TENORM) concerns and management approaches. TENORM contamination is typically the result of process operations involving the extraction, purification, filtration, smelting, or pipeline transport of virtually any material of geological origin.¹ The TENORM operations and activities discussed in this paper are not regulated by the Nuclear Regulatory Commission (NRC). We also do not discuss TENORM or naturally occurring radioactive materials (NORM) covered in the U.S. Environmental Protection Agency's (EPA's) Uranium Mill Tailings Regulations or covered by NRC's definition of "source material". What remains for discussion are radiological materials that are not federally regulated and are incidentally concentrated from various industrial processes, such as coal mining and combustion, and water treatment, and TENORM found in reused and recycled materials. These materials have the potential to be transported to solid waste facilities or become legacy contaminants on Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) sites.

Fate and transport analysis and risk assessment of TENORM to human and ecological receptors may reveal concerns inadequately reflected by its regulation. TENORM can drive risk and influence CERCLA remedial alternatives. Regulation is not addressed by the NRC and not by all States. Many State officials are left with managing legacy sites, Resource Conservation and Recovery Act (RCRA) C and D disposal facilities, and combustion wastes with essentially unregulated TENORM. But TENORM issues can be recognized early before they become real problems if solid waste management officials study TENORM. This paper is a good place to start.

2.0 INTRODUCTION

NORM is present in the environment; in soils, air and water. Industrial processes can separate and concentrate this material into TENORM, which is largely unregulated at the State and federal level. CRCPD has defined TENORM as meaning naturally occurring materials not regulated under the Atomic Energy Act of 1954, as amended, whose radionuclide concentrations have been increased by or as a result of human practices. TENORM does not include the natural radioactivity of rocks or soils, or background radiation, but instead refers to materials whose radioactivity is technologically enhanced by controllable practices or by past human practices. TENORM risk is based on concentrating the NORM thereby increasing the radiation exposure risk.

¹ Conference of Radiation Control Program Directors (CRCPD). *Part N - Regulation and Licensing of TENORM. SSRCR Volume I.* April 2004. <u>http://www.crcpd.org/SSRCRs/N_04-04-print.pdf</u>

TENORM is commonly associated with specific industries and practices. Examples include uranium mining and overburden, phosphate waste, coal waste, petroleum production scale and sludge, drinking water treatment, mineral mining/overburden and processing/extraction, and geothermal wastes. TENORM is primarily associated with NORM decay chains of uranium-238 and thorium-232 and their progeny (see Appendix B). Radium and radon are the main risk drivers in these decay chains. TENORM can present serious health and safety hazards if it is not handled and disposed of properly. Most producers of TENORM are not required to have a radioactive materials license and may not have the radiological expertise necessary to deal with the myriad of TENORM waste streams.

The issues related to TENORM storage and disposal are far reaching and expansive. It is neither possible nor feasible to attempt to address every issue in one document. Therefore, the ASTSWMO Radiation Focus Group will limit the scope of this paper to address TENORM issues which could potentially lead to unauthorized disposal of unregulated TENORM in solid waste landfills. This paper will include a review of TENORM basics and definitions, followed by sections devoted to toxicity and health effects; regulatory guidance; TENORM waste generation, extraction and disposal in various industries; and legacy sites. This guidance concludes with a focused case study on incidental TENORM in energy production and appendixes that provide basic information on radiation and decay chains for common NORM.

3.0 THE TENORM DILEMMA

TENORM consists primarily of material containing potassium-40 (K-40) and isotopes belonging to the naturally occurring radioactive primordial series. The principal radionuclides are isotopes of high atomic number and mass number elements belonging to the radioactive series headed by the three long-lived isotopes uranium-238 (uranium or U series), uranium-235 (actinium series), and thorium-232 (thorium or Th series). All three of these series have numerous radioactive species in their decay chains before reaching a stable isotope of lead.

In recent years, it has been recognized that certain materials used or generated in certain industry sectors have higher than average levels of NORM, or may concentrate the NORM into TENORM. As noted in a National Academy of Sciences report on NORM and TENORM, there are many industries that utilize materials containing NORM, or that concentrate NORM. Examples of industry sectors and the related NORM or TENORM material(s) that are largely unregulated are noted in the Table 1 below.

Industrial Sector	NORM or TENORM	Radioactive Series
Uranium mining	Waste overburden or low grade ore	U, Th
Other metal mining	Waste overburden	U, Th
Metal ore processing	Slag and sludges	U, Th

Table 1Industry Sectors and Associated NORM or TENORM

Industrial Sector	NORM or TENORM	Radioactive Series
Metal welding and	Metal alloys and products	Th
fabrication		
Metal casting, grinding or	Foundry sands or casting molds, grinding or shot	U <i>,</i> Th
sand-blasting	with zircons	
Phosphate fertilizer and	Waste phosphogypsum scale, residuals, slag	Ra-226, U
phosphorus production		
Oil and gas production	Process brine water treatment sludge, scale in	Ra-226, U, Th
	equipment, storage tank bottom sludge, gas	
	refining separation process	
Geothermal energy	Brine residuals and scale	Ra-226, U, Th
generation		
Drinking water treatment	Sludge and ion exchange resins	Ra-226, U, Th
Waste water treatment	Sludge	Ra-226, U, Th
Paper and pulp production	Scale and sludge	Ra-226, U, Th
Coal combustion for	Bottom and fly ash	Ra-226, U, Th
energy generation		
Decorative or optical glass	Slag and coating residuals	U, Th
Stone cutting and	Certain base rock (e.g., granite) with high U/Th	U <i>,</i> Th
polishing	series	
Building materials	Certain construction materials (e.g. gypsum and	Ra-226, U, Th
	stone (e.g. granite)	
Chemical industry and use	Potassium compounds	K-40

While regulations vary from State to State, operations involving NORM or TENORM are rarely licensed by the NRC or an equivalent Agreement State program. This is due to the fact that workers are unlikely to exceed 10 percent of the occupational whole body dose limit, which would require the use of a radiation monitoring device. There is very limited data, if any, on the industrial worker exposure from operations involving the generation of TENORM. In addition, TENORM may be of concern under non-occupational exposure scenarios. For example, TENORM radiation may exceed the cleanup criteria for CERCLA cleanups. For waste disposal, the incidental receipt of TENORM at RCRA C and D solid waste facilities may be in violation of the facility permit.

4.0 EXPOSURE, TOXICITY, AND HEALTH EFFECTS

Radiation dose to humans from NORM and TENORM is largely driven by radium isotopes from the U-238 and Th-230 decay chains. For external exposure pathways, Ra-226 and -228 are primarily responsible for external gamma exposure dose rates. Ingestion and especially inhalation of uranium and thorium decay products are of great concern because of the documented medical carcinogenic causes and effects to human health. Internalizing alpha emitters can pose significant human health effects from high alpha decay energies, long halflives, high dose conversion factors (slope factors), and high radiation quality factors. Radon gas and re-suspension of contaminated dirt or dust, especially particles sizes in the micron range, are of concern when evaluating radiation dose and risk assessment from alpha particle incorporation, which in most cases will involve bone deposition.

Drinking elevated levels of naturally occurring radionuclides in water or eating food grown in contaminated soils is the most likely path of ingestion. Inside the body, 85% of radium will end up in the skeletal structure where it will largely remain indefinitely. Radon's progeny (Po-218 and Bi-214) will impact the lungs and is the leading cause of lung cancer in non smokers. In addition to the risk from radiation exposure, uranium's prime consequence is attributed to chemical toxicity to the kidneys rather than radiological consequence.

For uranium mining, milling, or other source material operations, NRC or authorized Agreement States will have regulatory authority, and as such, have promulgated standards for exposure to radon compatible with 10 CFR Part 20. The Mine Safety and Health Administration (MSHA) also has jurisdiction for radon exposure monitoring in 30 CFR Part 57. Occupational Safety and Health Administration (OSHA) would have regulatory authority for radon exposure under 29 CFR Part 1910 in other work environs. OSHA incorporated the radon standard of ca-1970 NRC's concentration limits of 100 pCi L⁻¹ (3,700 Bq m⁻³). The current NRC standard for exposure to radon in 10 CFR Part 20, is 30 pCi L⁻¹ (1,100 Bq m⁻³).

Outside of uranium or other mining operations, there are no strict national building codes that require evaluation of occupational exposure to radon decay products. In recent years, a number of states and/or school districts have begun evaluating the radon levels in schools. It has been suggested that within a given radon zone, statistical significant correlation of residential and non-residential buildings requiring mitigation should be similar. U.S. EPA recommends a private dwelling residence be remediated when an indoor radon concentration exceeds 4 pCi L⁻¹ (148 Bq m⁻³). Given that OSHA's standards have not been updated, the National Council on Radiation Protection & Measurements (NCRP) recommends that indoor radon concentrations in most workplaces be limited to 30 pCi L⁻¹ (1,100 Bq m⁻³). In the workplace if there are sensitive populations exposed (e.g. in schools), NCRP recommends U.S. EPA's 4 pCi L⁻¹ (148 Bq m⁻³) concentration limit be applied. U.S. EPA's 0.02 WL UMTRCA standard is a potential Applicable or Relevant and Appropriate Requirement (ARAR) to consider for CERCLA remediations.

For additional information on the toxicity and health effects of the common NORM discussed in this section, please see:

- Agency for Toxic Substances and Disease Registry (ATSDR). <u>Radium ToxFAQs</u>. July 1999
- ATSDR. <u>*Radon ToxFAQs.*</u> September 2008
- ATSDR. <u>Uranium ToxFAQs</u>. September 1999
- ATSDR. Thorium ToxFAQs. July 1999

5.0 FEDERAL AND STATE REGULATORY GUIDELINES FOR MANAGEMENT OF TENORM

There are no uniform national guidelines or regulations for management of TENORM. Management of TENORM falls under various regulatory authorities including U.S. EPA, NRC, and the authority of individual States.

<u>U.S. EPA</u>

U.S. EPA regulates releases of TENORM to air from the phosphate industry and uranium mines through the Clean Air Act (CAA). It also regulates the liquid discharges of TENORM into surface waters from uranium mines and mills through the Clean Water Act (CWA). Regulatory oversight of vanadium operations are also through the Clean Water Act (CWA). The Safe Water Drinking Act (SWDA) protects community drinking water systems from TENORM.

U.S. EPA has the authority to clean up abandoned hazardous waste sites which may be contaminated with TENORM through CERCLA. The EPA has developed guidelines for cleaning up under the CERCLA remedial program media (e.g, soils, water, buildings) contaminated with TENORM and other radionuclides. which can be found at: http://www.epa.gov/superfund/health/contaminants/radiation/index.htm. Information on other EPA regulations and guidance regarding TENORM can be found at: http://www.epa.gov/rpdweb00/tenorm/regs.html

<u>NRC</u>

NRC regulates TENORM when concentrations of uranium and thorium are greater than or equal to 0.05% of the chemical mixture, compound, solution or alloy. At these concentrations, the mixture would be regulated as source material. Additionally, the Energy Policy Act of 2005 expanded the definition of byproduct material to include discrete sources of radium, which gave NRC the authority to regulate these sources. Additional information about the NRC's regulations can be found at <u>http://www.nrc.gov/materials.html</u>.

<u>States</u>

The following States have regulations covering TENORM: Arkansas, Florida, Georgia, Louisiana, Maine, Michigan, Minnesota, Mississippi, New Jersey, New Mexico, Ohio, Oregon, South Carolina, Texas and West Virginia. Some of these regulations are based on the suggested state regulations (SSR) developed by CRCPD. The CRCPD developed SSR Part N as a model standard guidance for regulating TENORM in an attempt to lead the states to uniform regulation of TENORM. CRCPD Part N materials can be downloaded at <u>http://www.crcpd.org/SSRCRs/N 04-04-print.pdf</u> and <u>http://www.crcpd.org/SSRCRs/Implement-Guide-print.pdf</u>

In March 2009, the American National Standard Institute (ANSI) and the Health Physics Society (HPS) approved and published a standard titled *Control and Release of Technologically Enhanced Naturally Occurring Radioactive Material* which specifies dose limits and release criteria for materials containing TENORM. ANSI/HPS N13_53-2009 is available through the Health Physics Society at <u>www.hps.org</u>.

6.0 TENORM WASTE PRODUCTION AND GENERATION

As stated above, there are many industries that produce wastes containing TENORM by concentrating the NORM in their processes. This section provides additional information about two of the most prominent of these processes: mineral extraction and drinking water treatment. This section also illustrates TENORM in building materials to show an example of end products of TENORM generation.

Mineral Extraction

Many mineral extraction processes create TENORM wastes including phosphate extraction, uranium mining and other metal and mineral extraction processes.

Phosphate extraction

The phosphate extraction industry is concentrated in the southeastern U.S. with Florida, North Carolina, and Tennessee being the largest producing states. Naturally occurring uranium, as well as its progeny, can be found in phosphate deposits. Uranium concentrations in phosphate ores found in the U.S. range from 7 to 100 pCi/g (0.25 to 3.7 Bq/l). Most phosphate rock is turned into fertilizer, but prior to being turned into fertilizer or other products it is transformed into either phosphoric acid (through the wet process) or elemental phosphorus (through the thermal process). This processing concentrates NORM in the waste products, transforming them into TENORM.

The primary waste byproduct of the wet-acid process to create phosphoric acid is phosphogypsum. Phosphogypsum contains appreciable quantities of NORM. Around 80 percent of the radium-226 becomes concentrated in the phosphogypsum. Radium concentrations at phosphogypsum stacks range from 11 to 35 pCi/g (0.4-1.3 Bq/g). Radon-222 can be found emanating from the surface of phosphogypsum stacks. Average radon fluxes range from 1.7 to 12 pCi/m2-sec (0.6-0.4 Bq/M2-sec) and can be as high as 340 pCi/m2-sec (13 Bq/m2-sec), with a mean value of 6.8 pCi/m2-sec (0.3 Bq/m2-sec).

The primary waste byproduct of the thermal process to create elemental phosphorus is phosphate slag. Phosphate slag is a glassy substance created during furnace processing. Phosphate slag's physical properties and high carbonate content renders slag to become less susceptible to radionuclide leaching than phosphogypsum. However, concentrations of uranium, thorium, and radium in phosphate slag have been measured as high as 50 pCi/g (1.85

Bq/g) in some instances. U.S. EPA has additional information on phosphate extraction at <u>http://www.epa.gov/radiation/tenorm/fertilizer.html</u>.

Uranium mining

The mining of uranium ores, by underground and by surface methods, produces small and large amounts of bulk waste material. This bulk waste material can be classified as TENORM. These TENORM inventories will include mining overburden and waste rock as well as evaporation pond sludges and scales.

These materials typically contain radionuclides of radium, uranium, and thorium. Uranium overburden contains very low levels of radionuclides. Radioactivity levels in in-situ leachate evaporation ponds are between 3 pCi/g (0.1 Bq/g) to 200 pCi/g (7.4 Bq/g). However, other solid TENORM wastes generated in uranium mining can reach radioactivity levels between 300 pCi/g (11.1 Bq/g) and 3000 pCi/g (111 Bq/g).

U.S. EPA has completed detailed technical reports from studies on uranium mining TENORM wastes which can be found on their website at http://www.epa.gov/radiation/tenorm/uranium.html.

Drinking Water Treatment Residuals

In 2003, the U.S. EPA Radionuclides Rule came into effect. The new rule required Community Water Systems to comply with maximum contaminant levels (MCLs) for NORM such as uranium, thorium and radium. Water systems in certain areas of the country have high levels of NORM and have had to implement additional treatment options to remove the radionuclides from the water.

In large water treatment plants, alum (aluminum sulfate), ferric chloride, or other chemicals are added to raw water, forming a gel that gradually coagulates or flocculates to the bottom of the tank as solid waste. Radionuclides and other impurities precipitate along with the solids with relatively high efficiency. Some type of sedimentation and filtration usually follows coagulation. A variety of filter media are available for use after coagulation in combination with fluoridation and chlorination. Activated carbon, sand, diatomaceous earth, greensand and membrane filters are all used. These filters may be backwashed and/or regenerated a number of times, but must ultimately be replaced. Most likely, smaller systems treat radionuclides and other contaminants with technologies, such as reverse osmosis (RO), ion exchange (IX) or softening. These treatment technologies generate solid waste as resins and membranes that will contain elevated levels of TENORM. These are likely generated in small amounts.

Drinking water treatment plant residuals are categorized as resins, membranes, filter media, sludges and liquids. Liquid residuals include brines, concentrates, backwash water, rinse water, and acid neutralization solutions. Spent resins, filters and membranes generally contain elevated levels of radioactivity that must be sent to a facility licensed for disposal of radioactive

materials. A specific radioactive materials license may be required due to elevated radioactivity. Facilities should conduct planning and modeling to avoid activity levels (particularly of radium) that will require disposal in licensed radioactive waste disposal sites.

Further information regarding management of TENORM for drinking water treatment facilities can be found in the EPA's A Regulators' Guide to the Management of Radioactive Residuals from Drinking Water Treatment Technologies: http://www.epa.gov/rpdweb00/docs/tenorm/816-r-05-004.pdf

Building Materials

Examples of building materials that may contain natural gypsum are mold resistance products, acoustically enhance gypsum, cement board, ceiling systems, joint treatment/finishing products, veneer plaster, etc. In the United States, U.S. EPA does not regulate NORM that might be present in building and construction materials, such as radium and thorium in natural gypsum. EPA does have regulatory oversight on phosphogypsum (PG)—mainly central/northern Florida and Idaho—as part of the air toxic program that precludes its use in building materials. PG has been used extensively in cement, wallboard, and other materials in Europe, Japan, and Australia. Potential users of PG might be in areas where it is more readily available than mined natural gypsum. There does not seem to be an abundance of published literature on radiation doses and airborne radon concentrations in actual structures built with PG building materials. In 1993, the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) estimated that for a residential building in which 4.2 tonnes of PG were used in plasterboard, the absorbed dose rate in air would approximately (7 μ Gy/ hr or 0.7mR/hr) and an effective dose rate from inhalation of radon progeny of about (0.6 mSv /yr or 60 mrem/yr).

As the popularity of granite countertops has grown in the last decade, the demand for them has increased tremendously. And with increased sales volume and variety, there have been more reports of "radioactive" or potentially hazardous countertops, particularly among more exotic and striated varieties from Brazil and Namibia. Most health physicists and radiation experts agree that most granite countertops emit radiation and radon at low levels. It is not uncommon for federal and state regulatory entities to receive calls from radon inspectors and home owners about granite countertops with radiation measurements several times above background levels. Most academic professionals in the U.S. have stated that the cancer risk from granite countertops in on the order of one in a million.

7.0 TENORM DISPOSAL

There are numerous categories of disposal facilities that may receive solid/sludge residuals containing TENORM; however, each increasing level of protection comes with increased costs and regulatory oversight. Examples are municipal solid waste landfills (MSWL), monofills,

industrial landfills, hazardous waste landfills, uranium mill tailings impoundments, and commercial low-level waste disposal sites.

American National Standards Institute/Health Physics Society (ANSI/HPS) Standard N13_53-2009 provides a framework for evaluating whether a solid or hazardous waste disposal facility should be allowed to accept wastes containing TENORM. The exposure and dose criteria used in the ANSI Standard apply to individual members of the public exposed to TENORM sources of radioactivity and radiation. Dose limits shall be expressed as total effective dose equivalent (TEDE). The criteria are:

(i) an annual dose limit of 100 mrem (1 mSv), above background, from all pathways and sources of radioactivity (except radon and its short-lived decay products); and practices associated with site and facility operations, including effluent discharges into the environment.

(ii) specific dose constraints for each practice and source of radioactivity, if established by an agency with legislative authority, shall be considered to ensure that dose limits are not exceeded when all individual dose contributions are added in determining the total dose.

(iii) an annual dose limit of 25 mrem (0.25 mSv), above background, from all appropriate pathways associated with the presence of residual sources of radioactivity (except radon and its short-lived decay products) from land and facilities that have been remediated and released for any use with no further radiological restrictions.

(iv) dose limits for internal and external exposures exclude doses from radon gases (219Rn, 220Rn, 222Rn) and its short-lived decay products. Exposures and doses to due to long-lived radon gas particulate decay products shall be included in the dose limit of Section 2.2.1.a(i). Sections 2.2.3 and 2.2.4 present criteria for exposure to indoor and outdoor radon gas, respectively, including short-lived particulate decay products from radon gas.²

(v) the Maximum Contaminant Levels (MCLs) for site and facility effluent discharges that may have an impact on current or potential sources of groundwater using the National Primary Drinking Water regulations, as amended, for the presence of 226Ra and 228Ra (combined), uranium, and gross alpha particle radioactivity (excluding radon).

To meet these exposure and dose criteria, the following enhanced facility controls may need to be implemented for acceptance of TENORM at solid or hazardous waste facilities:

• For planning purposes, no more than 1-10% of the volume of the cell should contain these subject materials.

² Sections 2.2.X refer to sections within ANSI/HPS N13_53-2009.

- The subject materials should be disposed of in a discrete area and covered immediately. A civil survey should be performed, preferably by a licensed professional surveyor for record keeping purposes. If GPS is used to locate the areas, the data should be differentially corrected to within 1m. No subject materials should be disposed within 3 meters of the final repository cover.
- The disposal facilities should conduct appropriate training of workers.
- The disposal facilities should employ dust control as necessary during staging and application of the materials, but may not add free liquids to a disposal cell.
- The facility should have a liner and leachate collection and recovery system.
- Leachate samples should be collected and analyzed for gross alpha, beta and gamma activity on an annual basis for screening. The data will be evaluated using trend analysis tools to determine if the solids residuals have affected the leachate. Should the screening evaluation indicate the potential release of alpha, beta or gamma activity into the leachate, then spectral analysis of leachate will be required.
- Additional groundwater monitoring requirements may be incorporated into the facility D & O Plan predicated on the above analytical results and data evaluation.
- If the facility does not have a leachate collection system, then the groundwater monitoring wells should be sampled and analyzed for speciated radioactivity.
- If the facility does not have groundwater monitoring wells, then an appropriate groundwater monitoring network should be established.
- Waste Acceptance: The landfill's approved waste acceptance plan should contain specific protocols for handling water treatment plant residuals. These will require the generator to ensure the residuals contain no free liquids as demonstrated by use of the paint filter test (U.S. EPA Method 9095) prior to disposal. Likewise, pH of the residuals should not be below 6.0 for landfill disposal. Finally, the generator should make a hazardous waste determination (based either on generator process knowledge or analytical results) to document the sludge does not fail toxicity characteristic for metals.
- Institutional controls and/or an Environmental Covenant should be required at closure of the solid waste disposal site and facility.

8.0 TENORM RECYCLING AND REUSE

The formation of TENORM containing scale in metallic objects from energy production, mining, and other operations can produce exposure pathways to workers and to a lesser degree consumers. Uranium series progeny (radium) deposited on the inside surfaces of pipes that are to be reused or recycled could potentially produce significant radiation doses. One scenario is when pipe from energy production may be cut up, rethreaded and reused as pipe). The facility and workers where the machining is done can be exposed and/or contaminated. The final user of the pipe may be exposed externally to gamma radiation or internally to beta/alpha radiation if the pipe is used for potable water. Finally, any other product moving through the remanufactured pipe may be contaminated depending on radio-chemistry. The International Atomic Energy Agency has a publication that describes the radiation hazards of gas and oil production.

The remelting of metal payloads with TENORM are often screened and detected by metal recyclers who employ truck portal radiation monitors to prevent sealed sources and TENORM from entering their facilities. The energy production industry is aware of this and they often clean metals before shipment to recyclers or for reuse). Be aware that even benign levels of radioactive materials can be rejected by metal recyclers. When a load sets off an alarm, the metal recycler usually refuses receipt of it. Repeated incidences will likely end recycling as a metal disposition alternative.

Most TENORM from metal production winds up in the slag byproduct (impurities skimmed off the top of molten metal). Some TENORM can hold up in air effluent treatment equipment (baghouse). The reuse of this TENORM containing waste in construction materials such as bricks, blocks, and concrete aggregate should be identified separately from background radiation to the general public. The metal product that goes on to consumers is typically clean since the TENORM partitions out with the slag.

A useful computer program for evaluation of metal recycling is RESRAD-Recycle, developed by Argonne National Laboratory. A default parameter computer run with 500 pCi/g (18.5 Bq/g) of Ra-226 in scrap metal partitions 99% of the radium to slag, 1% to the air filtration baghouse, and 0% to the metal ingot. Worker, transportation, and consumer scenarios can be evaluated. These partitioning percentages do not apply to all radionuclides.

Slag from phosphate production is also used for road building materials and contains TENORM. Coal ash contains variable amounts of TENORM and is sometimes used in construction materials such as concrete and road surfaces. Generally reuse scenarios do not result in significant radiation dose to the public. TENORM in residential building materials or native stone used in residential property provide the largest potential doses. Still, NORM in native geology with dark shale associated regolith under houses nearly always presents the greater potential dose through radon exposure.

Brine byproduct waters from oil and gas drilling can be recycled for use during dust suppression on roads and disturbed terrain on drill sites, pumping sites and service roads. TENORM in brine waters (ancient groundwaters as old as the sedimentary rocks in which they reside) varies greatly according to the organic material content of the geologic formations. Some brines are low enough in TENORM that they can be used for a variety of purposes on site. Knowledgeable studies of the particular drilling sites and geology may provide insights into the potential for presence of TENORM in scales and the brines, and the need for analytical sampling (SW846 lab methods for alpha counting will produce underestimates for high brine content waters). Usually scales, brine handling sludge treatments, and concentrated NORM and TENORM are more of a concern radiologically than the brine water itself. Note that untreated release of brines to surface waters may exceed water quality criteria for saline constituents if not TENORM. Reuse of brine (i.e. dust suppression) should consider the impact of all constituents on surface waters and aquatic organisms. Sludges from municipal and industrial wastewater treatment plants contain TENORM. The concentration of concern depends on the recycle or reuse scenarios. Use of sludge as an agricultural soil amendment presents the highest potential for human TENORM exposure. Characterization of the sludge and confident estimating of TENORM activities allow site specific analysis using the RESRAD modeling program. Consult appropriate specialists to insure the RESRADs input parameters are adequately conservative. Adjust them as appropriate for the specific site. Be mindful that municipal sludge can also contain radionuclides from hospitals or industry. Stormwater influx to sewers can carry environmental contaminants that end up in sludge. The RESRADs program will correct for half lives, ingrowth, and mobility in the environment. One may find that application of a sludge, as a soil amendment, has no contribution to native soil activities and dose. Or one may find it contributes about the same or less as store bought fertilizers. Or one may determine the sludge in question should be regulated by application rate or not used in agriculture at all.

Dose or risk associated with TENORM is not to be associated with normal background radiation. The NCRP, in Report 160 (2009), *Ionizing Radiation Exposure of the Population of the United States,* details recognized sources of background radiation and respective doses. The report documents an increase in background radiation mainly from medical tests and better characterization of other sources. Careful analysis of TENORM related risk and dose will assure that recycling does not add to the background recognized as part of consumer products and building materials. The need for distinction between NORM and TENORM has been recognized by EPA in testimony to Congress.

9.0 LEGACY SITES

Legacy sites may have never been properly characterized for all hazards including radiological components. The long term environmental monitoring of these sites may not include requirements for monitoring of radiological components due to not recognizing the potential a particular industry may have for generating TENORM wastes. Even current remedial projects may not recognize the potential for TENORM contamination due to personnel not being familiar with industries associated with TENORM generation.

TENORM associated with uranium, thorium, and their associated decay series is of the most concern. Some of these processes may generate source material, which is regulated by specific licensure by the NRC or NRC Agreement States. Source material is defined as any combination of thorium and uranium, in any physical or chemical form, or ores that contain by weight one-twentieth of one percent (0.05 percent) or more of uranium, thorium, or any combination thereof. Examples include water treatment resins and brine processing by the oil and gas industry that contain one-twentieth of one percent or more by weight of source material. Note that depleted uranium (left over from uranium enrichment) is considered source material. Some States have regulations specific to TENORM. In other states, regulations for the control of radioactive material implicitly include TENORM. TENORM occurrences are evaluated on a case by case basis to decide if a radioactive materials license or other controls are required for

possession or disposal of the material. These safeguards are "as low as (is) reasonably achievable" (ALARA) principles to minimize exposure to the public and workers.

10.0 FATE AND TRANSPORT

Fate and transport analysis and risk and dose assessment of TENORM to human and ecological receptors can reveal concerns that are inadequately reflected by its regulation. The biogeochemical behavior in a given decay chain can be expected to vary with atomic number. For example, in the uranium decay chain, isotopes of uranium, thorium, radium, radon and other elements occur in disequilibrium. Usually, they range from an inert gas to a readily absorbable, tetravalent cation of thorium. These properties determine the fate of the radionuclide in fuel and mineral processing, their transport in soil or surface disposal environments, and ultimately their biologic availability and uptake. Knowledge of their behavior is essential for defining source terms and assessing dose.

Regulations for controlling exposure of the public to radionuclides are often dose based. Usually, radiation doses result from interaction of humans with these radionuclides contained in environmental media, i.e. air, water, soil, and biota. A comprehensive working knowledge of the behavior of TENORM species in these media is essential. It is important to know:

- The different mobilities of various radionuclides in the decay chains.
- How technologic processes have changed the physical and chemical form of the radionuclides and the release rates of these species to the various media.
- Weathering reactions over time.
- The concentration, physical, and chemical form of the decay chain members.

General guidance on dose limits and release criteria for waste containing TENORM radioactive materials are outlined in the ANSI/HPS Standard N13.53-2009. The predominant radioactive species and progenies are associated with uranium 238 and thorium 232 decay chains. This ANSI Standard is applicable to industries and activities that are not regulated by federal and state regulations. The standard focuses on practices and operations that might amass and relocate radioactivity or make radioactivity more accessible to public. Protective measures and corrective actions might be critical to protect public health and safety. Activities and practices under this TENORM guidance are: mining and beneficiation of ores, processing of ore materials, gangue, feedstock used in the manufacturing of consumer and industrial products, and distribution of products containing TENORM. This ANSI/HPS Standard does not apply to NORM found in natural soils and rocks, and materials associated with common practices, i.e., scientific/research studies, engineering studies, soil/geologic sampling, farming, construction grading, grave digging, trench/excavation work, etc.

Doses and risks associated with TENORM are not unique and parallel with those associated with radioactive materials administratively regulated by U.S. EPA, NRC, Department of Energy (DOE), and Agreement States. Also, recommendations made by NCRP, International Commission on

Radiological Protection (ICRP), European Commission (EC), and Canadian Guidelines for NORM are appropriate guidance in establishing radiation protection principles for TENORM.

HPS recommends that the current version of DOE's Argonne National Laboratory RESRAD computer code be utilized to assess impacts of near surface disposal of wastes containing low-activity materials. RESRAD uses pathway analysis to calculate site specific parameters related to radiation doses and health risks. It is important to consult appropriate specialists to insure the RESRADs input parameters are adequately conservative. Adjust them as appropriate for the specific site. The RESRADs program will correct for half lives, ingrowth, and mobility in the environment. The State of Colorado illustrates how RESRAD can be used to make risk informed decisions regarding TENORM disposal its *Interim Policy and Guidance Pending Rulemaking for Control & Deposition of TENORM Materials* (February, 2007). To this end, a risk-informed approach should be applied to TENORM site assessments.

The economics of TENORM disposal provide an incentive to allow its disposal in non-hazardous waste landfills that also receive municipal solid waste. TENORM is now receiving more attention for its potential to cause elevated radiation exposures to the public.

11.0 SITE CHARACTERIZATION AND CERCLA DECISION MAKING

At first, historical negative repercussion from TENORM production was an unknown phenonema. Environmental surveillance of sites/operations revealed the presence of radioactive materials that necessitated further investigation. Once qualitative and quantitative measurements document the presence of TENORM materials, several issues should be considered in implementing a data quality objective (DQO) process:

Step 1. State the Problem: define the problem that necessitates the study; identify the planning team, examine budget, schedule

Step 2. Identify the Goal of the Study: state how environmental data will be used in meeting objectives and solving the problem, identify study questions, define alternative outcomes

Step 3. Identify Information Inputs: identify data & information needed to answer study questions

Step 4. Define the Boundaries of the Study: specify the target population and characteristics of interest, define spatial and temporal limits, scale of inference

Step 5. Develop the Analytic Approach: define the parameter of interest, specify the type of inference, and develop the logic for drawing conclusions from findings

Step 6. Specify Performance or Acceptance Criteria: specify probability limits for false rejection and false acceptance decision errors; develop performance criteria for new data being collected or acceptable criteria

Step 7. Develop the Plan for Obtaining Data. Select the resource-effective sampling and analysis plan that meets the performance criteria

The U.S. EPA has developed guidance for systematic planning using the DQO process. Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM) is a widely accepted final status survey and verification approach that is useful for buildings and shallow soil. MARSSIM is a graded approach in that not all areas of the site will have the same potential for residual contamination, thus, not all areas require the same level of survey. As regulators you should review and approve all survey plans. EPA does not recommend any particular methodology for final status surveys but do recommend the Soil Screening Guidance for Radionuclides for site characterization. It is critical to use the DQO process to identify data gaps that may be inherent to a method.

In response to the success of MARSSIM, Kentucky, California, and several federal agencies developed a supplement document: the Multi-Agency Radiological Laboratory Analytical Protocols Manual (MARLAP). MARLAP provides uniform guidance for the planning, implementation, and assessment phases of projects that require the laboratory analysis of radionuclides. In addition, the Multi-Agency Radiation Survey and Assessment of Materials and Equipment Manual (MARSAME), also a supplement to MARSSIM, has recently been released by the U.S. EPA, DOD, DOE, and NRC. MARSAME provides information on planning, conducting, evaluating, and documenting environmental radiological surveys to determine proper disposition of radioactive materials and equipment. Prior to MARSAME, the NRC Regulation Guide 1.86, "Termination of Operating Licenses for Nuclear Reactors," was typically used as the standard for surveying material and equipment for release. Copies of all guidance materials and additional information on the topics discussed above can be located at the following websites:

- Guidance on Systematic Planning Using the Data Quality Objectives (DQO) Process (EPA QA/G-4) http://www.epa.gov/QUALITY/qs-docs/g4-final.pdf
- Multi-Agency Radiation Survey and Site Investigation Manual (MARSSIM)
 <u>http://www.epa.gov/rpdweb00/marssim/obtain.html</u>
- Multi-Agency Radiological Laboratory Analytical Protocols Manual (MARLAP)
 <u>http://www.epa.gov/rpdweb00/marlap/manual.html</u>
- Multi-Agency Radiation Survey and Assessment of Materials and Equipment Manual (MARSAME)

http://www.epa.gov/rpdweb00/marssim/marsame.html

- NRC Regulation Guide 1.86 "Termination of Operating Licenses for Nuclear Reactors" <u>http://www.nrc.gov/reading-rm/doc-collections/reg-guides/power-</u> <u>reactors/active/01-086/01-086.pdf</u>
- Additional information on MARSSIM:
 - http://www.marssim.com http://www.epa.gov/radiation/marssim/

Establishing background can be an issue on sites, both for NORM, and for anthropogenic isotopes that we as regulators would consider "background", but not supported by environmental groups or the public. It is important to discriminate contamination originating from the responsible party from ubiquitous background distributions of radioisotopes, such as fallout and primordial radionuclides. The radionuclides of interest are identified with the site as a source of radioactive contamination. Make this clear in the DQO and in professional and public information.

The measurement of gross alpha and gross beta is common on sites because it is relatively simple and inexpensive. However, it is important that these results are properly interpreted. The responsible party might try to screen out environmental media entirely based on gross counts. This should be validated by isotopic analysis for an agreeable number of quality assurance (QA) samples. It could be that specific radionuclides such as tritium might be present that would not contribute to gross beta counts. Gross counts might not find the fringe of an unknown source term where isotopic analysis would.

Considering the limitations, gross counts can be used for screening groundwater that is suspected of being impacted by radioactive contamination. The probability of missing particular radionuclides must be considered in the DOQs. This screening method assumes that the groundwater is a potential source of drinking water. The screening process is based on U.S. EPA MCL guidance for radionuclides in groundwater. The primary analytical method for this screening is by gross alpha/beta counting via gas proportional counter (EPA method 900.0). If the gross alpha results exceed 5 ρ Ci/L, then the sample shall be analyzed for isotopic radium (Ra-226/Ra-228) constituents. If the gross alpha results exceed 15 ρ Ci/L, then the sample shall be analyzed for isotopic uranium (U-238/U-235/U-234) constituents.

If the initial analytical results (gross alpha) are not explained by the isotopic radium or uranium and daughters results, then perform alpha spectroscopy to determine other potential contaminants. Should the gross beta results exceed 50 pCi/L, then the sample shall be analyzed for strontium-90, or, if strontium is not a contaminant of concern, another potential beta emitter suggested by the historical site assessment. The sample shall be analyzed for tritium and compared to the tritium MCL of 20,000 pCi/L, most likely by liquid scintillation counting. Information on groundwater sampling can be found at:

http://www.epa.gov/safewater/radionuclides/regulation.html

Additional testing can be determined based on screening results, site history and process knowledge. The site investigation process can be iterative. The sampling plan and data objectives should recognize the probability of developing additional concerns and questions. The resolution is based on additional statistical sampling, site research, and refinement of remediation alternatives. Biased sampling can be used in an educated manner but cannot be used to prove a negative. The "If it isn't here, it isn't anywhere" statement cannot be statistically validated. It may be possible to obtain specific manufacture's documentation on site related radioactive materials and prove a positive however.

The following resources contain additional guidance and training for measuring radionuclides.

- ITRC Real Time Measurement of Radionuclides in Soil: Technology and Case Studies Guidance: <u>http://www.itrcweb.org/Documents/RAD_4Web.pdf</u> Training: <u>http://www.clu-in.org/conf/itrc/radsrealtime_102808/</u>
- U.S. EPA Soil Screening Guidance for Radionuclides (Current Site Characterization Methodology: <u>http://www.epa.gov/superfund/health/contaminants/radiation/radssg.htm</u>

Analytical Methods

For both potassium-40 and radium-226 and other gamma emitters within these decay chains, identification and quantification is easy. Most laboratories use U.S. EPA's Method 901.1. For the alpha emitting radionuclides, there exists very accurate alpha spectroscopy analysis which identifies specific isotopes such as Isotopic Uranium series (U-234, U-235 and U-238) and Isotopic Thorium series (Th-228, Th-230 and Th-232) radionuclides. For these isotopic analysis, laboratories use the American Society for Testing and Materials (ASTM) Method number ASTM D3972-90M. In waters with high dissolved solids, such as brines from deep wells, alternate alpha counting and spectroscopy methods might be needed that minimize counting signal attenuation. Table 2 provides a reference for analytical methods.

Table 2Radiological Methods for the Analysis of Soil/Sediment Samples

Parameter	Original Method	Instrument	Reference	Method #	MDA/MDL
	Evaporation and	Alpha/Beta Prop			
Gross alpha/beta	planchet	cntg	APHA, 1995	AB-001/B03	X=6pCi/g B=12pCi/g
Uranium - total	Evaporate and dissolve	KPA	ASTM, 1995	D-5174	0.1 ug/g
Uranium,	Separation &				
Isotopic	Microprecipitation	Alpha Spectroscopy	DOE 1990	AS-001/A03	0.1pCi/g
D 000	Preparation and		ED4 4000	004	
Ra-228	Separation	Beta Prop cntg	EPA 1980	904	1.0pCi/g
To 00	Preparation and ion	Liquid Scintilation	EII 1002	TC001	2.0 pCi/c
10-99	Ion Exchange &	Chily	LII, 1995	10001	2.0 pc//g
Thorium, Isotopic	Microprecipitation	Alpha Spectroscopy	1986/DOF 1990	AS-001/A03	0.1pCi/g/isotope
	Ion Exchange &		LANL.		0
Americium-241	Microprecipitation	Alpha Spectroscopy	1986/DOE,1990	AS-001/A03	0.1pCi/g
	Separation & Radon	Alpha Scintilation			
Ra-226	Demanation	Cntg	EPA 1980	903.1	0.1pCi/g
	Sample in Gamma	Gamma			
Bi-207	Counter	Spectroscopy	EPA 1980	901.1	see Gamma scan
Dh040/D:040	Solvent Extraction &	Data Dran anta	DOF 4000		2 = 0:/=
PD210/BI210	Precipitations	Beta Prop chtg	DOE, 1990	PD-055	3 pCI/g
Colhalt Isotonic	Counter	Spectroscopy	EPA 1980	901 1	see Gamma scan
	Preparation and LSC	Liquid Scintilation	LIA 1300	501.1	See Gamma Scam
Tritium	Cocktail	Cntg	EPA 1980	906	50pCi/a
		Gamma			
Gamma Scan:	Gamma Spectroscopy	Spectroscopy	EPA 1980	901.1	15 pCi/g
K-40					based on Cs137
Cs-137					u
TI-208					п
Pb-212					н
Bi-214					н
Ac 227/228					н
AG-221/220	(gamma spec) 21 Day				
Radium 226	ingrowth				"
	Separation plus Radon	Alpha Scintillation			
Radium 226	Emanation	cntg	EPA 1980	903.1	0.3 pCi/g
Radium 228	Separations	Beta Prop cntg	EPA, 1980	904	1 pCi/g
Isotopic,	Ion Exchange &				
Plutonium	Microprecipitation	Alpha Spectroscopy	DOE 1990	AS-001/A03	0.1 pCi/g
Total Sr	Separations	Beta Prop cntg	EPA 1980	905	1 pCi/g
Strontium 90	Separations	Beta Prop cntg	EPA 1980	905	3 pCi/a
Thorium 230					• • • • •
(Iso)	see Thorium, Isotopic				
Plutonium 238					
(Iso)	see Isotopic Plutonium				
		Gamma			
Protactium	Gamma Spectroscopy	Spectroscopy	EPA 1980	901.1	see Gamma scan
238/PA234	Gamma Spectroscopy	Spectroscopy	FPA 1980	901 1	0.2pCi/g
			D- 00	D- 001/	4 = 0'/=
P0-210	Preparation and Plating	DOE 1990	P0-02	P0-03W	i pui/g

MDA/MDL - Minimum Detectable Activity/Minimum Detection Limit (will vary with laboratory and count time) pCi/g - picocuries per gram

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STUDY: TENORM ASSOCIATED WITH ENERGY PRODUCTION

INTRODUCTION

Subterranean resources utilized for energy production include coal, oil, natural gas, and geothermal energy. Coal and oil/natural gas are part of the carbon based fuel cycle; whereas, geothermal is considered a renewable resource. These four resources are used for energy production by various industries. Each resource needs to be extracted from the earth, and, in the case of the carbon based fuels, burned to produce energy. The steps in the fuel cycle for each resource is examined for the presence of naturally occurring radiological materials (NORM) and whether the concentrations of these radionuclides may be anthropogenically enhanced to a point where their use or disposal may pose a risk human health and the environment.

COAL

Coal is used world-wide for the production of electricity, and as fuel for industrial boilers and other processes. Coal is primarily composed of organic matter (carbon) but also includes inorganic impurities or trace elements that do not readily combust. Prior to use coal must be extracted from geologic deposits and transported to utilities. The utilities then burn the coal to produce energy and leave behind coal ash that could have elevated levels of technologically-enhanced, naturally-occurring radioactive materials (TENORM) that will eventually be disposed in landfills or used in consumer products.

<u>Mining</u>

Coal is basically mined in two different ways: surface mining and underground mining. The choice of how the coal is mined is based on the depth of the coal deposits. For deep deposits, typically hard coal (anthracite), underground mining is used to extract coal. Associated with the extraction of coal from underground mines is produced water or coalbed brine. Brine is typically pumped from the mine to a sedimentation basin. TENORM may be present in the form of scale or sludge.

Gas, oil and coal mining operations generate large volumes of produced water. Produced water or brine, is often a highly contaminated effluent... (Tait, n.d.).

Chau & Chrus'ciell in 2007 observed sludge measurements collected from coalbed brine reservoirs in Poland indicated elevated concentrations of radium-226 (Ra-226), and radium-228 (Ra-228). The reported concentrations were 2,500 picocuries per gram (pCi/g) and 47.8 pCi/g for Ra-226; and 1,236 pCi/g and 70.5 pCi/g for Ra-228. Each of the two concentrations is from different reservoirs with different chemistries. These concentrations can be compared to

typical soil background concentrations for Ra-226 of approximately 0.7 pCi/g. Uranium did not appear to be mobilized with the brine solutions.

Sediments associated with the discharge of coal bed brines, as well as the brines themselves should be considered as potential sources of TENORM. Other radionuclides to be considered during the mining of coal may be the unintentional release of radon and its daughters in dusts from air cleaning systems.

Coal Combustion

There are four major types of coal used to produce energy. These include lignite, subbituminous, bituminous, and anthracite coal; listed by increasing hardness and carbon content. The carbon content of coal ranges from 25 – 95 percent. The remaining inorganic materials contain trace elements, including NORM. *Most coal contains uranium and thorium, as well as potassium-40, lead-210 (Pb-210), and Ra-226. The total levels are generally about the same as in other rocks of the Earth's crust.* (NORM 2009)

Although the quantity of NORM in different types of coal varies, the range found in all coal is from below (0.3 - 1.4 pCi/g). Similar uranium concentrations are found in a variety of common rocks and soils. (FS-163-97, 1997) Figure 1 indicates the typical range of uranium concentration in a variety of materials.



Typical range of uranium concentration in coal, fly ash, and a variety of common rocks.

Figure 1. Graph from Radioactive Elements in Coal and Fly Ash: Abundance, Forms, and Environmental Significance. U.S. Geological Survey Fact Sheet FS-163-97. October, 1997

A study performed by the Department of Energy Environmental Measurement Laboratory (DOE-EML) (Beck & Miller, 1980) further indicates that the uranium-238 (U-238) and thorium-232 (Th-232) concentrations of coal used in U.S. power plants is comparable to the concentration found in typical U.S. soils. Table 1 shows the typical concentrations of U-238 and Th-232 in U.S. coals used at power plants and soils.

	Nuclide:	U-238	Th-232
Samples collected from mines	Mean	0.60	0.50
	Range	< 0.1 - 15	< 0.1 - 5.3
Samples collected from power	Mean	0.58	0.24
plants and analyzed by EML	Range	0.13 – 1.8	< 0.1 - 0.47
Other reported analysis of	Mean	0.34	0.56
samples from power plants	Range	< 0.1 - 0.8	0.17 – 2.3
Soil (for comparison)	Mean	0.70	0.70
	Range	0.3 - 1.4	0.2 - 1.3

*Values for mine samples are for coal as mined, for power plants for air-dried samples.

The majority of the carbon and organic constituents are burned during the combustion of coal. The unburned coal and trace element residue is called coal ash, which includes fly ash and bottom ash. Coal ash contains NORM as well as silicon, aluminum, iron, and calcium; these elements make up about 80 to 90 percent of all of the constituents of coal ash. Approximately 90 weight percent of coal is burned during the combustion process leaving 10 weight percent coal ash. *The concentration of most radioactive elements in solid combustion wastes will be approximately 10 times the concentration in the original coal.*

During coal combustion most of the uranium, thorium, and their decay products are released from the original coal matrix and are distributed between the gas phase and solid combustion products. The partitioning between gas and solid is controlled by the volatility and chemistry of the individual elements. Virtually 100 percent of the radon gas present in feed coal is transferred to the gas phase and is lost in stack emissions. In contrast, less volatile elements such as thorium, uranium, and the majority of their decay products are almost entirely retained in the solid combustion wastes. Modern power plants can recover greater than 99.5 percent of the solid combustion wastes. (FS-163-97, 1997).

The radionuclides that are part of coal ash are the same as those present in coal; including uranium, thorium, and their radioactive decay products including radium. The amount of radium in coal can vary by more than two orders of magnitude depending upon the type of coal and where it was mined.

Coal ash is composed of three different types: fly ash, bottom ash, and boiler slag. Fly ash is entrained with hot flue gases, trapped by stack filters, and

accounts for about 74% of the ash generated. Bottom ash, too large or heavy to be entrained, settles to the bottom of the boiler and accounts for about 20% of the ash generated. Boiler slag, formed when the ash melts under the intense heat, collects at the bottom of the boiler and in exhaust stack filters, and accounts for about 6% of the ash generated. (Coal Ash , 2009)

Tables 2 and 3 indicate typical radiation levels in coal ash from two different sources.

Wastes	Radiation Level [pCi/g]			
	low	average	high	
Bottom Ash	1.6	3.5-4.6	7.7	
Fly Ash	2	5.8	9.7	

Table 2. Typica	l radiation	levels in coal	ash (Coa	l Ash, 2009)
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Table 3. Radioactivity in Coal Ash, pCi/g (Naturally Occurring Radioactive Material (NORM), 20
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				Uranium series,	
Source		Total activity	Pb-210	Ra-226	Thorium series
UNSCEAR	world	98.5 pCi/g			
Gabbard	USA	32 pCi/g			
CSIRO	Australia	71.1 pCi/g			
	NSW	86 pCi/g			
Cooper 2003	Australia	up to 55.9 pCi/g			
Cooper 2005	NSW		3.5 – 5.4 pCi/g	2.2 – 4.1 pCi/g	2.7 – 5.4 pCi/g
	Victoria		0.41 pCi/g	0.54 pCi/g	0.41 pCi/g
	Queensland		1.1 – 2.7 pCi/g	1.9 – 3.2 pCi/g	1.4 – 4.3 pCi/g
Earth's crust	world	38 pCi/g			

Stack filtration devices, such as electrostatic precipitators, baghouses, and scrubbers are routinely used to reduce the emission of fly ash to the atmosphere by at least 95 percent. A small fraction of the fly ash produced, typically 2-5 percent, is released into the air (Coal Ash, 2009).

According to U.S. EPA, typically 70 to 80 percent of coal ash is disposed of in dry landfills. A landfill for a typical coal fired power plant (500-1000 Megawatts) requires about 74 to 148 acres. These landfills range from about 10 to 197 acres, and may be as much as 30 feet deep. (Coal Ash, 2009) It is estimated there are 305 off-site coal-ash landfills and surface impoundments and that there are about 900 on-site disposal facilities in the United States.

(Industrial Sectors with TENORM, 2009) This is over 200 square miles of land used for landfill disposal of coal ash.

The remaining coal ash (roughly 58 million metric tons per year) is beneficially used as additives in a variety of applications. Fly Ash is used in concrete and cement and in concrete blocks. It is typically substituted for cement in concrete at about 10-30%. It is also used as filler for asphalt. Sludge, is used in wallboard, as roadbase, and other miscellaneous applications. Boiler Slag is used for blasting grits and roof granules. Bottom Ash may be used for snow and ice control and other miscellaneous applications (ASTSWMO, 2009; NORM, 2009).

Since the early 1970s, all three types of coal ash have been used in construction projects. Coal ash is used to level out uneven terrain or applied as a stable fill for building construction. Typical applications include sites where shopping malls, housing developments, and industrial parks are planned for construction. Other projects have included the construction of road embankments, runways, public transportation system structures, and soil stabilization.

Other emerging applications of fly ash include the construction and sinking of artificial reefs, metal (aluminum and iron) extraction via direct acid leaching, and as filler in paints and plastics. Examples of products which may contain fly ash include paints and undercoatings, auto bodies and boat hulls, PVC pipes, battery cases, bowling balls, utensils and tool handles, vinyl floor covering, and shower stalls.

(NORM, 2009)

The radiological footprint from the use of coal to produce power is wide ranging. The redistribution of NORM is seen from the leaching of mining production water to the use of coal ash in construction and consumer products. The radiological dose from the coal fuel cycle has been estimated by DOE, U.S. EPA, and the U.S. Geological Survey to be on the order 10 μ Sv (2 mrem), which is far less than our exposure to background radiation.

The risks due to exposure to radionuclides from utilities are substantially lower than the risks due to natural background radiation. The average exposure to natural background radiation (excluding radon) for the U.S. population has been estimated to be roughly 1 mSv per year (100 mrem per year), which is about 67 times higher than the highest exposure due to utility radionuclide emissions. (Technology Transfer Network, 1998).

Caution needs to be exercised when evaluating the local exposure to NORM from the coal fuel cycle. The wide variation in measured concentrations of uranium, thorium, and radium in coal ash can, in some cases, exceed risk-based clean-up values commonly used at remediation sites.

OIL AND GAS PRODUCTION

Oil and gas production begins with the extraction of the resource, crude oil or gas, from subterranean geologic deposits to the surface. The processing of the raw resource may produce TENORM in piping, tanks and equipment. Petroleum industry sites may have released sludges, scales, and produced water to surface soils. Environmental investigations should be initiated because of the possibility that petroleum industry sites may be contaminated with TENORM.

Since NORM and oil are present in subsurface geologic formations, it is likely that during the extraction of oil that some NORM will be extracted with the oil. *Radionuclides are known to be associated with organic materials in nature; therefore, oil, gas, and oil field brines frequently contain radioactive materials.* (NORM Fact Sheet Naturally Occurring Radioactive Materials, 1997) The primary radionuclides present in these geologic formations are the isotopes U-238, Th-232, and their progeny. The primary radionuclide of concern is Ra-226. Ra-226 concentrations can range from undetectable levels to 40,000 pCi/g. (Saint-Fort, Alboiu, & Hettiaratchi, 2007)

Studies suggest that NORM is limited to the piping and tanks used to remove and process petroleum products and NOT in finished petroleum products (Al-Saleh & Al-Harshan, 2008). Tables 4 and 5 show the result of sampling for NORM in both petroleum products and petroleum wastes. (Note that Ra-224 is considered to be in secular equilibrium with Ra-228). The results of this sampling effort support the suggestion that NORM is enhanced in the residues from process piping and tanks during the processing of petroleum products. The finished petroleum products do not appear to be enhanced in NORM.

Ra-226, Pb-210, and Ra-228 are the primary radionuclides of concern in the petroleum industry. Ra-226 and Pb-210 are deposited as scale in pipes and equipment.

The waste streams most likely to be contaminated by elevated radium concentrations include (i) produced water, which is formation water that is brought to the surface along with the produced oil and gas; (ii) scale, which is a hard, insoluble deposit that accumulates on the surfaces of equipment and solid debris that come in contact with produced water; and (iii) sludge, which is a slightly granular, usually hydrocarbon-rich deposit that accumulates in the bottom of some storage and process vessels. (Smith, Arnish, Willliams, & Blunt, 2003)

Radium is slightly soluble and present in groundwater. The radium is extracted to the surface with the produced water (oil field brines) stream. The radium content in produced waters varies geographically and by geologic formation. When the produced water is brought to the surface some of the dissolved radium may precipitate. This precipitate will have elevated concentrations of radium. Radium concentrations tend to be highest closest to the wellhead where changes in temperature, pressure, and pH are greatest. The radium that does not

precipitate is typically disposed of with the produced water stream. A common way of disposing produced water in the United States is via subsurface injection. The radium content of the injected water is not regulated and may cause elevated radium content in the groundwater. Radium precipitation is not a problem in well scale formation.

Periodically, the scales and sludges that accumulate inside pieces of oilfield equipment are removed. Radium-bearing scales and sludges pose a waste management issue if the radium content exceeds specified exemption levels. Similarly, pieces of equipment containing residual quantities of TENORM-bearing scales and sludges and surface soils impacted by these wastes can present a waste management problem.

(Smith, Arnish, Willliams, & Blunt, 2003)

Published data (quoted in Cooper 2003) show radionuclide concentrations in scales up to 300,000 Bq/kg (8100 pCi/g) for Pb-210, 250,000 Bq/kg (6800 pCi/g) for Ra-226 and 100,000 Bq/kg (2700 pCi/g) for Ra-228. In Cooper 2005, the latter two maxima are 100,000 and 40,000 respectively. (NORM, 2009)

The front end of the petroleum fuel cycle appears to have considerable potential for the presence of TENORM. Petroleum industry sites, especially oil fields, should be suspected of having potentially elevated levels of NORM.

TENORM IN MARCELLUS SHALE GAS PRODUCTION

Marcellus Shale (MS) is found under significant areas of New York, Pennsylvania, Ohio and West Virginia, and is believed to be one of the largest gas reserves in the world. However, MS contains elevated levels of NORM associated with the uranium (U) and thorium (Th) present in the formation. The U content has been noted to be in the 10 to 100 parts per million (ppm) range. The natural radioactive decay of U / Th over time leads to the formation of other radionuclides such as Ra-226 and Ra-228. (PaDCNR, 2008)

All the radioactive elements present in MS can be a potential source of radiation exposure. For example, gas well drilling operations can bring MS rock cuttings with NORM to the surface. External gamma radiation dose rates from these cuttings have been found to be similar to other rocks that contain elevated U / Th (e.g., granite); typically less than 20 micro-roentgen per hour (uR/h).

The development of a MS gas well involves very large volumes (i.e., over a million gallons) of high pressure hydraulic fracturing, or "fracing," fluid. This process dissolves some of the materials bound-up in the shale formation such as salt, organics and other stable elements. Some of this water returns or flows back to the surface following the fracing of the well, and can potentially contain technologically enhanced NORM (TENORM) from the shale formation. Samples of this flow back water indicate that soluble Ra-226 and other radionuclides are

present. Data from a PaDER study in the early 1990s and the recent NY state draft EIS on MS notes Ra-226 in brines from these deep MS wells can be on the order of thousands of picocuries per liter (pCi/L). (PaDER and NYS dEIS, 1995).

Given the high levels of total dissolved salts (TDS), organics and other materials in frac water, it may not be directly discharged to the environment. Thus, treatment of these wastewaters can further concentrate the Ra-226 and other radionuclides in the resultant treatment waste solids. The effluents from these wastewater treatment facilities may also have elevated Ra-226 and/or gross alpha activity, and should be monitored. External gamma dose rates around waste containers of sludge have been measured in the background to 150 uR/h range. Thus, the disposal of these sludges must comply with respective state regulations or national guidance for exposure to the public, and should not exceed 25 millirem per year (mrem/yr). Disposal options to approved landfills for these TENORM containing sludges vary, but should be routinely tracked and reported to the regulatory authority.

NATURAL GAS

The extraction of natural gas may have similar NORM considerations as oil wells. The drilling for natural gas may also involve the presence of production water which may contain radium. The potential for the precipitation of radium with elevated concentrations in production water pipes needs to be considered.

Radon is present in natural gas. Eisenbud (1987) reports the concentration of radon in natural gas can range from 40 pCi/L to greater than 1000 pCi/L. Natural gas is blended and processed to produce liquefied petroleum gas (LPG) which consists primarily of propane. The boiling point radon and propane are similar, so during processing radon concentrations are increased in LPG and decreased in the methane-rich gas used in pipelines.

Radon-222 long-lived daughters, lead-210 (Pb-210) and polonium-210 (Po-210), tend to accumulate on the interior surfaces of the LPG plant machinery and constitute a potential source of exposure of maintenance personnel. (Eisenbud, 1987)

The investigations of LPG plant machinery for the presence of Pb-210 and Po-210 in elevated concentrations need to be considered.

GEOTHERMAL ENERGY PRODUCTION WASTE

Geothermal energy is produced by pumping high-temperature fluids from the ground. In order to access the high-temperature fluids boreholes are drilled and pipes are inserted to transport the fluids to the surface. *Geothermal energy currently makes a relatively minor contribution to total U.S. energy production. The primary geothermal development sites in the U.S. are the Geysers, in Sonoma County in northern California, and the Imperial Valley in southern California.* (Geothermal Energy Production Wastes, 2009)

The high-temperature fluids are contained in subterranean reservoirs in geologic formations. The rock in the formations may contain minerals and NORM that may be in solution with the high-temperature fluids. The present NORM in geofluids are uranium, thorium, and their associated progeny. If radionuclides are present in solution, the mineral scale, production sludges, and waste water will contain TENORM. The primary radionuclides produced with the geothermal fluids are Ra-226 and Ra-228.

The saline solutions extracted from the geothermal reservoirs may have dissolved solids content approaching 30 percent by weight. *The only significant TENORM wastes from geothermal power production are the solid wastes originating from the treatment of spent brines such as in Imperial Valley* (Geothermal Energy Production Wastes, 2009). The average Ra-226 concentration for these wastes is estimated at 132 pCi/g. The range of Ra-226 concentrations is shown in Table 6.

Table 6. (Geothermal Energy Production Wastes, 2009)				
	Radiation Level (pCi/g)			
	Low	Average	High	
Geothermal Energy Waste Scales	10	132	254	

...the likely scenario for hot rock scenarios is that uranium, thorium, radium, radon, and lead isotopes will be present in circulating geofluid in concentrations similar to dilute, natural groundwaters. Although there may be scales and sludge forming in surface plant, these are unlikely to contain significant activities of radium, the main source of gamma radiation in residues from the oil and gas industry. However, until the radionuclide content of hot rock residues has been established, due care should be taken in their handling. In particular, inhalation of residue dusts should be avoided, since this may introduce ionizing radiation doses from alpha and beta emitting radionuclides. (Battye & Ashman, 2008)

In conclusion, the likelihood of encountering TENORM from the solid wastes associated with geothermal energy production is associated with the solids content of the geofluids. If the solids content of the geofluids is small then the likelihood of encountering Ra-226 as TENORM is small. Conversely, if the solids content is elevated, then the accumulation of Ra-226 as TENROM is likely.

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APPENDIX A: RADIATION BASICS

An unstable or radioactive nucleus will release excess energy by emitting particles or Electromagnetic radiation. The common forms of radiation that may be emitted include: alpha (α) particles, beta (β) particles, and gamma (γ) radiation or photons. Nuclei that produce nuclear radiation are considered radioactive.

An alpha particle is relatively massive compared to a beta particle and it has a +2 charge, which causes it to strongly interact with electrons as it passes thru matter. Alpha particles do not travel far in air and are stopped by the dead external layers of skin causing no damage to the skin. Internal exposure to alpha particles can be very damaging to internal organs due to their high kinetic energy and its deposition in a short range.

Beta particles are highly energized electrons with a single negative (-1) charge, or positrons (+1), that travel a short distance in air. Some beta particles have enough energy to penetrate the live thickness of skin, and if sufficient activity is present, can cause acute damage. Beta radiation can affect the lens of the eye causing cataracts at very high doses. Internal exposure from beta particles can cause damage to internal organs of the body.

Gamma rays are very penetrating and can be damaging to internal organs from outside the body. A radioactive nucleus (radionuclide) may go through a single to many steps or decay transitions until the nucleus reaches stability. This series of steps is called a decay chain. Individual nuclei decay at different rates. The time it takes for one half of a given population of nuclei to decay is called a half-life. Half-lives of different radionuclides can vary from very short times on the order of microseconds (1 x 10E-06 sec or millionths of a second) to billions of years. The shorter the half-life the more radioactive the material, and faster the radionuclide decays away. A radionuclide with a long half-life will take a long time to decay away, but will be less radioactive.

Unlike common chemical measurements that are based on mass or chemical reactivity, measurements of radioactivity are based on detection of radiations emitted from a substance (e.g., water, soil, air filter, etc.). The "activity" of a particular media is a measure of how often a particle or photon is emitted from the substance per unit time. Each time a nucleus emits a particle or photon, the nucleus has decayed or disintegrated. The rate (disintegrations in a unit of time) at which nuclei decay is how radioactivity or activity is measured. In the U.S. it is often customary to still use the old unit for activity, the curie (Ci). The international or "SI" unit of radioactivity is the becquerel (Bq), which is equal to a single disintegration per second (dps). There are 3.7×10^{10} dps (or Bq) in a curie. Activity of sources is typically stated in micro- or millicuries, μ Ci or mCi respectively. Environmental samples are often reported as activity in a unit mass or activity in a unit volume. Soil and sediment samples may be reported as picocuries per gram (pCi/gram), where a pCi = $1 \times 10-12$ Ci. (a millionth of a millionth Ci). Liquid samples may be reported as pCi/L and air samples may be reported as pCi/.

Conversion Factors

To convert from	То	Multiply by
Curies (Ci)	becquerels (Bq)	3.7 x 10 ¹⁰
millicuries (mCi)	megabecquerels (MBq)	37
microcuries (μCi)	megabecquerels (MBq)	0.037
millirads (mrad)	milligrays (mGy)	0.01
millirems (mrem)	microsieverts (μSv)	10
milliroentgens (mR)	microcoulombs/kilogram (µC/kg)	0.258
becquerels (Bq)	curies (Ci)	2.7 x 10 ⁻¹¹
megabecquerels (MBq)	millicuries (mCi)	0.027
megabecquerels (MBq)	microcuries (μCi)	27
milligrays (mGy)	millirads (mrad)	100
microsieverts (μSv)	millrems (mrem)	0.1
microcoulombs/kilogram (μC/kg)	milliroentgens (mR)	3.88

http://orise.orau.gov/reacts/guide/measure.htm

Dose

The amount of emitted energy actually absorbed or deposited in tissue, or other material, is called the radiation absorbed dose (rad). The units commonly used to quantify this dose are the rad (U.S. Customary) and the gray (Gy, SI units). The rad and Gy are measures of energy absorbed per unit mass of material. The relationship between rad and Gy is: 1 Gy = 100 rad. When radiation collides with living cells, a number of potential outcomes are possible. Some of these outcomes include no damage to the cell, death to the cell, and damage to the cell. Damage to a living cell depends on the amount of energy transmitted to the cell. The amount of energy transmitted to a cell depends on the type and energy of the radiation. Different types of radiation produce different effects and different levels of damage to cells. An alpha particle will interact more intensely and transfer energy over a shorter distance within a cell than a photon with the same energy. The alpha particle is more "effective" at producing biological damage than the photon, even though the total amount of energy transferred is identical overall. The same amount of absorbed dose from different types of radiation will have different biological effects. The Dose Equivalent (DE) is an expression of dose in terms of its biological effect. Appropriate weighting factors can be applied to the absorbed dose reflecting the different relative biological effects (RBE) to find the equivalent dose. The dose equivalent is expressed in units of rems (Roentgen Equivalent Man) or sieverts (Sv). The dose equivalent in rems is the radiation dose in rads multiplied by a weighting factor, which is an assessment of the relative biological effectiveness of that particular type and energy of radiation. Alpha particles have a weighting factor as high as 20, so that one rad is equivalent to 20 rems. For

Photons the quality factor is so that one rad and one rem are equivalent for gamma and x-rays. The conversion from sieverts to rem is: 1 Sv = 100 rem

Risk

Carcinogenic slope factors are used to assess risk from radionuclides just like for other chemicals. According to EPA Memorandum OSWER 9355.5-30, January 4, 2010, remedial Superfund cleanups generally should achieve a level of risk within the 10-4 to 10-6 carcinogenic risk range based on the reasonable maximum exposure for an individual.

The EPA's electronic calculators provides preliminary remediation goals for radionuclides. Per EPA policy, the calculator provides concentrations in both activity and mass units. EPA's recommended models for CERCLA sites are as follows: risk from radionuclides in soil and water (PRG), risk from radionuclides inside buildings (BRPG), risk from radionuclides on hard outside surfaces (SPRG), and complying with dose based ARARs (DCC). These calculators represent EPA's newest guidance and replace Risk Assessment Guidance for Superfund (RAGS) chapter 10 of Part A and chapter 4 of RAGS part B. Radiological risk should be considered cumulative to chemical risk. EPA's current site characterization methodology is found at: "Soil Screen Guidance for Radionuclides":

http://www.epa.gov/superfund/health/contaminants/radiation/radssg.htm.

One common radionuclide, uranium, has an acute chemical toxicity that is a concern for the ingestion pathway. If a site has uranium, this potential hazard should be noted and evaluated. The chemical toxicity of uranium can drive its risk as much as its carcinogenicity. Chemical toxicity of uranium in soil, water, or air should be assessed using the Regional Screening Level (RLS) electronic calculator for chemicals. Other resources for assessing risk from radionuclides are provided by the following:

• U.S. EPA - "EPA Preliminary Remediation Goals for Radionuclides" (PRG calculator) <u>http://epa-prgs.ornl.gov/radionuclides/</u>

U.S. EPA "EPA Preliminary Remediation Goals for Radionuclides in Buildings" (BPRG calculator) http://epa-bprg.ornl.gov/

U.S. EPA "EPA Preliminary Remediation Goals for Radionuclides in Surfaces" (SPRG calculator) http://epa-sprg.ornl.gov/

 U.S. EPA - "Risk Assessment Guidance for Superfund" <u>http://www.epa.gov/oswer/riskassessment/ragsa/index.htm</u>

- U.S. EPA "Radiation Risk Assessment at CERCLA Sites: Q & A" http://www.epa.gov/superfund/health/contaminants/radiation/pdfs/riskqa.pdf
- U.S. EPA "Superfund Radiation Risk Assessment and How You Can Help: An Overview" <u>http://www.epa.gov/superfund/health/contaminants/radiation/radvideo.htm</u>

APPENDIX B: PRINCIPAL NATURAL RADIONUCLIDE DECAY SERIES

Radionuclide	Symbol	Half-life	Major emissions
Uranium 238	²³⁸ U	4.47 x 10 ⁹ y	alpha
Thorium 234	²³⁴ Th	24.1 d	beta
Protactinium 234 IT (99.84% feeds 234U)	^{234m} Pa	1.17 m	beta, gamma
Protactinium 234 (0.16% feeds 234U)	²³⁴ Pa	6.7 h	beta, gamma
Uranium 234	²³⁴ U	2.46 x 10 ⁵ y	alpha
Thorium 230	²³⁰ Th	7.54 x 10 ⁴ y	alpha
Radium 226	²²⁶ Ra	1,600 y	alpha, gamma
Radon 222	²²² Rn	3.82 d	alpha
Polonium 218	²¹⁸ Po	3.10 m	alpha
Lead 214	²¹⁴ Pb	26.8 m	beta, gamma
Bismuth 214	²¹⁴ Bi	19.9 m	beta, gamma
Polonium 214	²¹⁴ Po	1.64 x 10 ⁻⁴ s	alpha
Lead 210	²¹⁰ Pb	22.3 y	beta, gamma
Bismuth 210	²¹⁰ Bi	5.0 d	beta
Polonium 210	²¹⁰ Po	138.4 d	alpha
Lead 206	²⁰⁶ Pb	stable	none

Table 1: Uranium Decay Series (including ²²⁶Ra)

Radionuclide	Symbol	Half-life	Major emissions
Thorium 232	²³² Th	1.41 x 10 ¹⁰ y	alpha
Radium 228	²²⁸ Ra	5.75 y	beta
Actinium 228	²²⁸ Ac	6.15 h	beta, gamma
Thorium 228	²²⁸ Th	1.91 y	alpha, gamma
Radium 224	²²⁴ Ra	3.66 d	alpha, gamma
Radon 220	²²⁰ Rn	55.6 s	alpha
Polonium 216	²¹⁶ Po	0.14 s	alpha
Lead 212	²¹² Pb	10.64 h	beta, gamma
Bismuth 212	²¹² Bi	1.01 h	alpha, gamma
Polonium 212 (64% from 212Bi)	²¹² Po	3.00 x 10 ⁻⁷ s	alpha
Thallium 208 (36% from 212Bi)	²⁰⁸ TI	3.05 m	beta, gamma
Lead 208	²⁰⁸ Pb	stable	none

Table 2: Thorium decay series (including ²²⁸Ra)