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Fate of Radionuclides In Wastewater Treatment Plants

A thesis submitted in partial satisfaction of the
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by

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ABSRACT OF THE THESIS

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In the western United States and in many arid regions, wastewater reclamation is becoming a common way of increasing water supplies. More and more wastewater is being reclaimed for non-potable uses such as irrigation, but reclamation for potable use is also being practiced. One of the concerns for wastewater reclamation is the distribution of contaminants that are not removed by either the wastewater treatment plant or the water treatment plant in the case of potable reclamation. The recent accident at Fukushima has focused attention on the spread of fission and decay byproduct across farmlands and into drinking water systems. An important concern is the distribution of radionuclides by wastewater reclamation. In former times, it was assumed that reclamation activities will stop if there is a contamination problem, but with increasing reliance of reclamation, stopping reclamation may have important and perhaps severe effects, including the loss of key industries such as petroleum refining that use reclaimed water. This thesis is a review of the important radionuclides that may be produced from a fission accident or a spill, and their fate in a wastewater treatment plant. Since experimental approaches for studying this problem are not generally feasible, a review of all the documented occurrences of spills has been made. In some cases the fate of fallout from atmospheric testing has been observed and this also provides useful information. The overall goal is to develop the knowledge that will allow treatment plant operators to react quickly to an accident or spill and provide guidance for temporary or short term measures to mitigate the impacts.

The thesis of Farzaneh Shabani Samgh Abadi is approved.

Keith D. Stolzenbach

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1. INTRODUCTION

Recent events have increased public interest in radionuclides in the environment. The Fukushima accident released approximately 150 x 10^{15} Bq ^{131}I and 50 x 10^{15} Bq ^{137}Cs into the environment (Yoshida and Kanda, 2011), but more importantly reminded the public of potential accidents and their environmental impact.

Radionuclides exist in our environment from many sources and reactor accidents are only one of the important sources. Local impacts of reactor accidents can be major as observed in the Ukraine and Belarus from the Chernobyl accident or minor as observed from the Three Mile Island Accident. The impacts of the Fukushima accident are not known at present but will likely be significant.

The impacts of these important accidents to the rest of the world are much reduced by comparison to the local impacts, and other sources may be more important. Public perception often tends to ignore sources that are not sensational and therefore do not attract the news media. For example, exposure to radon in basements or residuals from weapon testing has more impact on human health in the United States than any of the well-known reactor accidents.

A reactor accident in the United States could have large impact and its impact might be worsened by the transport of radionuclides though our water and biosolids recycling systems. Areas in California such as the Los Angeles Metropolitan area are heavily depend on water reclamation. Treatment plants in these areas often rely on beneficial reuse of biosolids for disposal. The inability to recycle water or reuse biosolids because of low-level radiological contamination will have major economic impacts.

The goal of this thesis is to review the sources of radionuclides with an emphasis on determining those that may impact water and biosolids reclamation. The second goal is to determine what temporary or "just in time" treatment solutions may exist or could be developed to mitigate the effects of a reactor accident.

Data documenting the distribution of radionuclides are rare but there are some observations of the fate of radionuclides from fallout, spills and hospital discharges that can be used to understand the likely fate of larger releases for an accident.

This thesis is arranged in eight chapters:

1. Introduction
2. Natural Radioactivity, which includes some information about naturally occurring radionuclides
3. Nuclear Power Plants, which includes some basic information about nuclear power plants and their performance

4. Radionuclides from Weapon Tests, as a considerable source of radionuclides in the environment
5. Radionuclides from Accidents, which includes information about radionuclides released to the environment due nuclear accidents
6. Radionuclides in Water Supplies
7. Radionuclides in Wastewater Treatment Plants, which summarizes information and data from different references about fate of radionuclides in wastewater treatment plants, and
8. Radionuclides from Medical Facilities, which represents information about medical facilities releases of radionuclides as a more permanent source of these materials.
9. Conclusions

2. NATURAL RADIOACTIVITY

Among 340 nuclides found in nature, 70 are known to be radioactive. Radionuclides in nature can be categorized in three groups based on their sources. The first group includes those with such long half-lives that they have not decayed since creation. These are called primordial. Members of this group have half-lives around 10^{10} years or more. An example of these types of nuclides is ^{40}K, which is known as the most important isotope causing human radiation exposure. The second group is from radioactive decay of primordial radionuclides, and the third group is called cosmic radionuclides because they are produced by bombardment of stable nuclides by naturally occurring neutrons formed by cosmic radiation. ^{3}H, ^{22}Na, ^{14}C and ^{7}Be are classified as the more important ones impacting human health.

Naturally occurring radionuclides can be further divided in to two groups: those that occur singly and those that are decay products of three series: the Uranium series starting with ^{238}U, the Thorium series starting with ^{232}Th and the actinium series starting with ^{235}U. A forth series, called the Neptunium series, was known to exist, but Neptunium and its decay products have disappeared almost completely due to their short half-lives. Neptunium now only exists if it is produced artificially. Members of each of these groups are shown in Tables 1-1 through 1-5.

Human external exposure from natural radioactivity is primarily from ^{40}K and the three decay series and less from the primordial isotopes and the cosmic group. The most important radionuclides of these groups are discussed in following sections.

Table 2-1: Radionuclides Induced in the Earth's Atmosphere by Cosmic Rays
From NRCP (1987)

Radionuclide	Half-life	Major Radiation
^{10}Be	1.6 x 10^6 yr	β
^{26}Al	7.2 x 10^5 yr	β^+
^{36}Cl	3 x 10^5 yr	β
^{80}Kr	2.13 x 10^5 yr	X ray
^{14}C	5.37 x 10^3 yr	β
^{32}Si	650 yr	β
^{39}Ar	269 yr	β
^{3}H	12.33 yr	β
^{22}Na	2.6 yr	β^+
^{35}S	87.4 d	β
^{7}Be	53.3 d	γ
^{37}Ar	35 d	X ray
^{33}P	25.3 d	β
^{32}P	14.28 d	β
^{38}Mg	21 hr	β
^{24}Na	15 hr	β
^{38}S	2.38 hr	β
^{31}Si	2.62 hr	β
^{18}F	109.8 m	β^+
^{39}Cl	56.2 m	β
^{38}Cl	37.29 m	β
^{34}mCl	31.99 m	β^+

Table 2-2: Non-Series Primordial Radionuclides
From NRCP (1987)

Radionuclide	Half-life (yr)	Major Radiation
^{40}K	1.26 x 10^{9}	β,ϒ
^{50}V	6 x 10^{15}	ϒ
^{87}Rb	4.8 x 10^{10}	β
^{113}Cd	> 1.3 x x10^{15}	Not Reported
^{115}In	6 x 10^{14}	β
^{123}Te	1.2 x 10^{13}	X rays
^{138}La	1.12 x 10^{11}	β, ϒ
^{142}Ce	>5 10^{16}	Not Reported
^{144}Nd	2.4 x x10^{15}	α
^{147}Sm	1.05 x 10^{11}	α
^{152}Gd	1.1 x 10^{14}	α
^{174}Hf	2.0 x 10^{15}	α
^{176}Lu	2.2 x 10^{10}	e^- , ϒ
^{187}Re	4.3 x 10^{10}	β
^{190}Pt	6.9 x 10^{11}	α
^{192}Pt	1 x 10^{15}	α
^{209}Bi	>2 x 10^{18}	α

Table 2-3: Uranium Series
From Bureau of Radiological Health (1970)

Radionuclide	Half-life	Major Radiation
^{238}U	4.47 x 10^{9} yr	α, <1% ϒ
^{234}Th	24.1 d	β, ϒ
^{234m}Pa	1.17 m	β, <1%ϒ
^{234}Pa	21.8 yr	β, ϒ
^{234}U	2.44 x 10^{5} yr	α, <1%ϒ
^{230}Th	7.7 x 10^{4} yr	α, <1%ϒ
^{226}Ra	1600 yr	α, ϒ
^{222}Rn	3.8 d	α, <1%ϒ
^{218}Po	3.05 m	α, <1%ϒ
^{214}Pb	26.8 m	β, ϒ
^{218}At	2 s	α, β
^{214}Bi	19.9 m	β, ϒ
^{214}Po	164 μs	α, <1%ϒ
^{210}Tl	1.3 m	β, ϒ
^{210}Pb	22.3 yr	β, ϒ
^{210}Bi	5.01 d	β
^{210}Po	138.4 d	α, <1%ϒ
^{206}Tl	4.20 m	β, <1%ϒ
^{206}Pb	Stable	None

Table 2-4: Thorium Series
From Bureau of Radiological Health (1970)

Radionuclide	Half-life	Major Radiation
232Th	1.4 x 1010 yr	α, <1%ϒ
228Ra	5.75 yr	β, <1%ϒ
228Ac	6.13 hr	β, ϒ
228Th	1.91 hr	α, ϒ
224Ra	3.66 d	α, ϒ
220Rn	55.6 s	α, <1%ϒ
216Po	0.15 s	α, <1%ϒ
212Pb	10.64 hr	β, ϒ
212Bi	60.55 m	α, ϒ
212Po	0.305 µs	α
208Tl	3.07 m	β, ϒ
208Pb	Stable	None

Table 2-5: Actinium Series
From Bureau of Radiological Health (1970)

Radionuclide	Half-life	Major Radiation
^{235}U	7.038 x 10^{8} yr	α, ϒ
^{231}Th	25.5 hr	β, ϒ
^{231}Pa	2.276 x 10^{4} yr	α, ϒ
^{227}Ac	21.77 yr	β, <1%ϒ
^{227}Th	18.72 yr	α, ϒ
^{223}Fr	21.8 m	β, ϒ
^{223}Ra	11.43 d	α, ϒ
^{219}Rn	3.96 s	α, ϒ
^{215}Po	1.78 ms	α, <1%ϒ
^{211}Pb	36.1 m	β, ϒ
^{215}At	0.1 ms	α, <1%ϒ
^{211}Bi	2.14 m	α, ϒ
^{211}Po	0.516 s	α, ϒ
^{207}Tl	4.77 m	β, <1%ϒ
^{207}Pb	Stable	None

2.1- Uranium

Natural uranium consists of three isotopes: ^{234}U, ^{235}U and ^{238}U. In the earth's crust Uranium is composed of 99.28% ^{238}U, 0.71% ^{235}U, and 0.0058% ^{234}U, which causes naturally occurring uranium to have specific radioactivity of 13 kBq/g.

With a specific gravity of 18.7, it is the heaviest of all naturally occurring elements. Uraninite (UO_2) and pitchblend (U_3O_8) are the most common uranium minerals found in nature.

Uranium can be found in rocks and soils around the world at different concentrations. Its concentration varies based on rock type and location, and ranges from 0.5-4.7 ppm in common rock types to as high as 120 ppm in phosphate rocks of Florida, which have been used as a commercial source of uranium. (NCRP,1993) The average concentration of uranium in soils is less than in typical rocks. Seawater also contains a noticeable amount of uranium with average concentration of 1.3 ppb, but it is not known if this source of uranium can be used commercially in near future.

Uranium is also present in air. Airborne soil and coal fly ashes are known to be possible sources of uranium in the air. In 1987, the National Council on Radiation Protection (NCRP) adopted the representative concentration of uranium in the air of the United States to be 0.7 $\mu Bq/m^3$. Uranium is also present in surface water and the intake from tap water varies by location. The concentrations of uranium in domestic waters of the United States may range from 0.4 Bq/m^3 to 130 Bq/m^3 in different states. Uranium can be found in phosphate rocks, and as a result in phosphate fertilizers. The presence of uranium in fertilizers and soils introduces it to the food chain and consequently into human tissues. Lung, kidney and bones receive the highest doses of uranium.

2.1.1- Uranium reserves and processing

Before and during World War II, uranium sources were known to exist in the Belgian Congo (Shinkolobwe mine), the Great Bear Lake of Canada, in the former Czechoslovakia (Joachimsthal) and in the Colorado plateaus. After World War II, new explorations for uranium sources began in the United States and other countries.

Uranium deposits found worldwide can be divided in to 14 different groups, among the most important ones are:

1) Unconformity related deposits constitute one third of the worldwide uranium resources. These are the main types of deposits found in Canada and Australia,
2) The Olympic Dam which is located in Australia and constitutes 66% of the Australia’s uranium sources,
3) Sandstone deposits, which constitute 18% of uranium resources worldwide. Major uranium sandstone states in the US include Wyoming, Colorado and Texas. These also can be mined in Niger, Kazakhstan, Uzbekistan and Gabon,
4) Surficial deposits, which constitute 4% of uranium resources worldwide,
5) Volcanic deposits, which constitute only a small portion of uranium resources and can be found in China, Kazakhstan, Russian Federation

and Mexico and

6) Quartz-pebble conglomerate deposits, which constitute 13% of uranium resources worldwide including those in Canada and South Africa.

Today's most important Uranium ore suppliers are Canada, Australia and Kazakhstan.

During the period between 1940 and 1970, most of the uranium mined was used for nuclear weapon production. However, the current usage of uranium is mostly for nuclear power plant fuel production. Conventional sources of uranium around the world can provide approximately 4.7×10^6 Mt of uranium, which, considering annual worldwide usage of 65,000 Mt, means these sources are a reliable supply for at least 70 years. Unconventional sources of uranium including phosphate deposits and ocean water are estimated to have the potential to provide 22×10^6 Mt and 4000×10^6 Mt uranium, respectively; however, these sources are still not considered economically feasible to mine.

Different technologies for uranium mining include underground and open pit mining. ISL (In Situ Leach) or solution mining is used to provide an increasing portion of world's uranium. This method is more environmental friendly than the other mining technologies; however, concerns about groundwater contamination are associated with this method. Radon and its decay products are the only significant gaseous products from that have been released from mines to the environment. The radioactivity of discharged air from mines to the environment range from 0.65 to 26 kBq/m^3, which is not greater than what is added by radon flux through 1 km^2 of earth surface and as a result they cause insignificant health risks. Controlling the ventilation rate of mines may control these releases.

In the process of preparing uranium, ores are sent from mines to mills where U_3O_8 concentrate, which is known as yellowcake, is produced. Milling process includes three steps: 1) ore grinding, 2) alkaline leaching and 3) uranium concentration by ion exchange or solvent extraction. During this process 95% of the uranium is removed; however, all the other radionuclides from uranium series remain in the mill tailings. Tailing piles can cause several environmental impacts mainly through radon emission by wind dispersion, leaching in water and being used as construction materials. As mentioned before all the other nuclides from the uranium series, except uranium itself will remain in tailings. These include ^{230}Th with 80,000 year half-life, which later decays to radium and consequently radon. As a result mill tailings will have radium and radon emission for thousands of years. According to US EPA report (1982), ^{226}Ra concentration in mill tailings ranges from 1.85 to 37 kBq/kg comparing to 0.04-0.08 kBq/kg in normal soils. Radon diffuses to piles surfaces and consequently to the atmosphere. Radon emission rate through this process is very sensitive to barometric pressure. 1% change in barometric pressure may cause 50-100 % change in radon emission rate to the atmosphere (Clements and Wilkening, 1974).

Tailing piles may cause releases to the environment due to impoundment failure and erosion by flowing water or wind action. Impoundment failure can be prevented by different engineering methods. Erosion can be controlled by covering milling tailings with rocks and by diverting floodwater. Asphalt, clay and cement are used as covers to reduce radon emission. New tailings can be protected from release to the environment by burying in shallow pits and locating them away from flooding regions. Underlying plastic or clay barriers can also provide ground water protection. However, the environmental impacts of tailing piles are localized and can be detected only within a few miles from these piles. Death risk due to radon emission is low even in the worst conditions. US EPA has set a limit of 0.74 $Bq/m^2/s$ for release from mill tailings over 1000 years; however, the expenditures typically required to meet this limit are high compared the health risk.

Liquid tailings have often been released to the nearby streams, which creates an elevated dose to wildlife and vegetation downstream of mills compared upstream of mills.

Mill concentrates are then sent to refineries, where it is converted to either metal or other uranium intermediate compound such as U_3O_8 or UF_4. The type of waste from uranium refineries depends on the material being processed. During World War II, when high-grade ores with as much as 4 MBq/kg ^{226}Ra were being processed, refinery sludge contained as much as 40 MBq/kg ^{226}Ra. However, today's concerns about uranium refineries are mostly associated with chemical wastes rather than radioactive wastes.

Uranium enrichment is the next step in the process of preparing uranium. Gaseous-diffusion enrichment used to be the predominant process for enrichment in the United States; however, laser enrichment is more common now. The by-product of enrichment process is called depleted uranium, which constitutes 89% of the original feed. Every ton of natural uranium being processed provides 130 kg of enriched fuel with more than 3.5 % ^{235}U.

Spent fuel may be recovered to separate ^{235}U and transuranic elements such as ^{239}Pu. If fuel recovery is planned, spent fuel must be stored in water-pools for weeks to months to let the fuel cool down and the short-lived fission products to decay. From World War II up to 1966, four governmental fuel-reprocessing facilities were in service in the United States with the major activity occurring at Hanford, WA and Savannah River, GA. They were mostly associated with the nuclear weapons program. The Oak Ridge facility was engaged in all sorts of nuclear programs including plutonium recovery and Idaho Falls facility mostly served navy and research reactors.

Environmental releases from fuel recovering plants may be caused by normal plant operation and plant accidents such as those that happened in Los Alamos, Hanford, Oak Ridge and Idaho. Also unprotected recovery of fuel during World

War II, due to urgent need for plutonium, caused leakage from storage tanks and ^{131}I release to the environment. However, releases are preventable by controlling the amount of fissionable materials being processed.

2.2- Radium

^{226}Ra belongs to uranium series with half-live of 1622 years and decays to ^{222}Rn through alpha emission. It does not have a gamma emission itself; however, its decay products are gamma emitters with stable ^{206}Pb, being the last decay product.

Radium can be found in rocks and soils in different concentrations ranging from 16 - 48 Bq/kg in typical rocks and it is nearly in equilibrium with the uranium in these sources. It is also present in surface and ground waters as the decay product of uranium. It has also been measured in different concentrations in public water supplies in the United States. In 1976, the NPIDWS set a limit of 185 Bq/m^3 for ^{226}Ra in drinking water supplies. Comparing radium concentrations with this limit around the United States reveals that approximately 75% of water supplies exceeded this limit, mainly in two regions: 1) piedmont and coastal areas of Atlantic ocean states and 2) Minnesota, Iowa, Missouri, Illinois and Wisconsin. Following these results, four different drinking water methods were studied for radium removal in Illinois and Iowa. Among these four methods, RO and sodium ion exchange were 92% efficient for radium removal. Lime-soda ash softening was 75 to 95% efficient and systems designed for iron removal were 11 to 53% efficient. Since radium concentration exceeded the US EPA limits by a wide margin, and the costs for treating radium are relatively high, new regulations were evaluated to reduce cost but still be protective of health.

Radium is present in soils and since it is chemically similar to calcium, it will be taken up by plants and enters the human body through the food chain. The ICRP (1973) reported that 70 to 95 % of radium ingested will deposit on bones.

2.3- Radon

Radium in rocks and soils decays through alpha emission to radon, and some fraction of it contaminates water in soils and pores of rocks, and may result in soil gas contamination. The contaminated soil gas may be exchanged to the atmosphere and as a result ^{222}Rn will be transferred to atmosphere. Another possibility for ^{222}Rn to enter the atmosphere is through diffusion from soil gas to atmosphere, which is less important compared to the exchange mechanism. Radon concentration in air is less than that in soils. Radon concentrations in typical soils ranges from 4 to 40 kBq/m^3, however, the average annual air concentration of radon in the United States ranged from 0.6 to 28 Bq/m^3. It is reported (Gesell, 1983) that the maximum concentration on ^{222}Rn is measured in fall and winter and the minimum concentration in spring, which is inconsistent with the atmospheric turbulence regime. Several factors affect the atmospheric radon concentration including air passage over the ocean and precipitation, distance from the surface of the earth and soil moisture. Radon and its decay products can also attach to the dust in the air and reach equilibrium. Radon decays through some short half-life radionuclides to ^{210}Pb, which has a half-life of 22.3 years. Equilibrium between radon and ^{210}Pb is reached in two hours in the air.

Radon may enter the human body by inhaling contaminated air, which allows this nuclide to enter the lungs. The Radon that decays in the lungs is no longer a gas, and the decay products remain in the lungs. The radiation dose received from ^{222}Rn itself by lungs is much less than that received from radon decay products.

Contaminated water is another source for human exposure to ^{222}Rn. Drinking water supplies in the United States have different concentrations of radon ranging from 3.7 to 960 kBq/m^3. The limit of 11.1 kBq/m^3 for radon set by US EPA (1991) were exceeded by many drinking water supplies and based on this limit, EPA estimated that 15 million people in the United States are served by water systems with doses higher than this limit. However, the human body can tolerate higher concentrations of radon in water and the US EPA is developing new limits.

Indoor air is another source of human exposure to radon and this type of exposure has gained scientific interests since the 1970s. Controlling indoor air ventilation rates, following efforts made for energy conservation, used to be known as the main factor causing high doses of radon indoor air contamination. Later it was understood that air ventilation rates do not play an important part in varying radon concentrations and other important factors such as pressure difference between indoor and outdoor, radon concentrations of soil and soil permeability impact the contamination. Radon concentrations in building materials are related to its concentration in rock fractures, soil pores and the permeability of the ground to the gases. Smaller soil pores and fewer rock fractures and more liquid in pores, tend to reduce radon content.

^{222}Rn enters indoor air by different methods. It diffuses from contaminated construction material. It also diffuses through foundations soil; however, this transfer mechanism is not comparable to radon transfer due to atmosphere air and soil air exchange. Another source is tap water, which does not add high doses, unless it is highly contaminated. ^{222}Rn concentration in the United States homes indoor air reported to have arithmetic mean value of 55 Bq/m^3 and geometric mean of 33 Bq/m^3. These values are consistent with what reported by US EPA (1992) showing 54 Bq/m^3 and 38 Bq/m^3 for arithmetic and geometric means respectively.

Several methods may be used to reduce indoor radon including crawl space ventilation, sub-slab suction, and sealing openings in the foundation. These methods might be used together to provide radon reduction. Providing filtration systems or fans supplemented with ion generators is also another method for radon reduction. Radon concentrations in water can be reduced by aeration or granular activated carbon.

The US EPA set 150 Bq/m^3 as allowable radon activation level and NCRP set the limit of 300 Bq/m^3. According to the US EPA, 5.8 million US homes have radon indoor doses higher than 150 Bq/m^3.

2.4- Potassium

^{40}K is the only radioactive isotope among three potassium isotopes. It decays to stable ^{40}Ar through beta and gamma emission. Natural potassium consists of 0.0118% radioactive potassium, causing it to have specific activity of 30 kBq/kg. Potassium contents of various types of rocks range from 0.3 to 4.5 percent. It is also present in seawater at 11 kBq/m^3 concentrations. It is estimated that 3000 Ci of potassium is added yearly to soils in the United States in the form of fertilizers. (Guimond, 1978); however, environmental levels do not influence the potassium content of human body and the dose of ^{40}K in human body is constant. It is the predominant radionuclide in foods and human body due to its energetic beta emissions and causes doses to gonad and other soft tissues and bones.

2.5- Phosphate fertilizers

As mentioned earlier, uranium is present in phosphate rocks and as a result in phosphate fertilizers. Uranium content of the ores in the United States range from

8 to 400 ppm. This radionuclide and its decay product radium tend to be separated from fertilizers during phosphoric acid production, which is an important step in fertilizer manufacturing. According to NCRP report (1993), depending on the price of uranium, it may be extracted as a side stream during phosphoric acid production. Phosphate fertilizers do not add high doses to the public; however, phosphate contaminated rocks may be present in cattle feed, which will cause contamination in cow's milk.

Two by products of phosphate fertilizer processing, gypsum and calcium silicate, are used as building materials. These may increase buildings gamma radiation dose. According to NCRP (1978), in the United States and Canada indoor gamma radiation equals that of outdoor, however, this may differ in some buildings.

2.6- Fossil Fuel

Coal contains radionuclides from uranium and thorium series and also ^{40}K. Coal uranium and thorium concentration in the Unites States are reported as1.7 μg/g and 4.5 μg/g respectively. The amount of radionuclides released due to coal combustion depends on factors such as radionuclides concentrations in coal, method of combustion and the efficiency of fly ash recovery. After combustion radionuclides appear in ashes. According to the NCRP, modern certified power plants achieve 99.5% ash retention. Emissions from coal-fired power stations do not affect uranium and thorium background compared to other sources.

Radionuclides also are present in petroleum gas and their release through oil and gas industry has been a recent subject of concern. Some states including Texas, Louisiana and Mississippi have regulations for natural radioactivity related to oil and gas industry.

3. NUCLEAR POWER PLANTS

The first nuclear reactor was operated at the University of Chicago in 1942, four years after discovery of nuclear fission. During the World War II, nuclear reactor technology developed rapidly. One year after Fermi and his associate's work at the University of Chicago, a 3.8 MWt reactor operated in Oak Ridge, TN, which remained in service for 20 years as radioisotope supplier. The first plutonium production reactor with initial power level of 250 MWt went critical in 1944 in Washington.

During the period of 1945 to 1954, the US nuclear program was under the complete control of the government and all the reactors were constructed within governmental sites and mostly used for military purposes. After World War II and the passage of Atomic Energy Act in 1954, which coincided with President Eisenhower Atoms for Peace program, private industry was allowed to enter the nuclear energy field and they began to develop nuclear power plants. The nuclear power industry enjoyed growing interest prior to the mid 1970s for several reasons, including:

1) Increasing need for electricity in the US by an average annual rate of 6%,
2) Possibility of building larger nuclear power plants, which made them more economically feasible,
3) Increasing concerns regarding air pollution associated with fossil fuel

power plants, and

4) Increasing price of the fossil fuel due to uncertainties in world politics.

The first commercial power reactor began operation in 1963 by the New Jersey Central Power Company. Up to the mid 1970s, nuclear power plants were built rapidly and orders for new ones were confirmed. However, during 1970s, several factors led to set back in nuclear power plant popularity, including:

1) Economical situation during 1970s,
2) High capital cost of nuclear power plants compared to that of fossil fuel plants,
3) Public concerns about nuclear power plants safety after the Three Mile Island (TMI) accident in 1979, and
4) Increased capital cost of nuclear power plants in order to meet safety requirements and waste disposal difficulties, especially after TMI accident

These were the main reasons leading to cancelation of all the orders for building new nuclear power plants after 1973. The Tennessee Valley Authority's Watts Bar I was completed in 1996, and is the last reactor approved in 1970's to begin operation.

Currently, 103 reactors are operating at 65 different sites in 31 states in the US. In six of these states, nuclear power plants provide more than 50% of electricity. Interests in building new nuclear power plants began to rise again after 2000 owing to several reasons including: high price of natural gas, improved performance of existing reactors, uncertainties about future restrictions on coal emissions and reduction in operating cost of nuclear generated electricity. Several orders for building new nuclear power plants have been approved and others are being evaluated. However, the Fukushima Daiichi nuclear accident in 2011 created new concerns and environmentalists have raised new doubts regarding nuclear power plants safety.

3.1- Nuclear Rector's Basics

In the US, power reactors use either natural uranium with 0.7% ^{235}U or enriched uranium up to 3%. Plutonium has also been used as reactor fuel. Reactor fuel can be prepared both as UO_2 or metallic uranium. However, there are some disadvantages associated with metallic uranium. It may swell after several neutron bombardments and it is very active chemically, and reacts exothermally with air and water. These are the reasons why UO_2, in spite of its poor heat conduction properties, is preferred over metallic uranium.

Reactor fuel is usually fabricated as rods, pins, plates or tubes and is protected by stainless steel, zirconium on other special claddings, in order to prevent the escape of fission products. The cladding also protects the fuel from erosion effect in case of contact with coolant. UO_2 has melting point of 2200° C and due to poor thermal conductivity, it is fabricated in 1.2 to 2 cm diameter rods. Normally UO_2 cylinders (pellets) with 1.2 to 2 cm diameter and 2.5 cm long are mounted in 1.3m long tubes, with helium filled spaces between them, which helps to enhance the heat exchange from fuel to cladding. These fuel tubes are arranged with control rods, which capture excessive neutrons in the reactor and are constructed of materials such as cadmium, indium, boron and hafnium, in gridded plates inside the reactor vessel.

During operation of nuclear reactors, each fission results in the multiplication in the number of neutrons. Some neutrons will escape from the reactor and non-fissionable materials inside the reactor will capture additional neutrons. A reactor is known to be self-sustainable when for every atom that fission, one fission-inducing neutron remains after capture by non-fissionable materials and leakage from the reactor. In this situation the reactor is called to be in critical condition.

The state of criticality of a reactor might also be expressed using a factor called neutron multiplication factor, *k*, which is the ratio of the number of neutrons produced by each fission to the number of neutrons generated in the preceding fission. The ideal situation for power reactors to operate is when k equals unity meaning that the reactor is in critical condition. Reactors with k less than unity are known as sub-critical and reactors with k greater than unity are known as supercritical.

The probability of fission depends on the energy of the neutrons. In most power reactors, thermal neutrons with energies as low as 0.1 eV increase the probability of fission. The energy of fission-causing neutrons can be controlled by materials such as water and graphite, which are called moderators.

Critical condition in a reactor is influenced by the presence of control rods, radiation history of the core and temperature. Increases in reactor temperature causes decreases in coolant density and as a result decreases moderation ability of the coolant and consequently decreases fission probability due to high energy levels of neutrons. Also increase in temperature of the reactor will increase the capture cross-section of the non-fissionable material in the reactor and decrease the number of fission-inducing neutrons inside the reactor. These entire effects cause negative temperature coefficient for the reactor, which is required for the reactor to be able to self regulate itself.

A good design for a nuclear reactor must consider nuclear, thermal and structural properties. As an example, choosing appropriate materials should minimize corrosion in the reactor. Corrosion materials may become radioactive due to neutron bombardment inside the reactor. ^{60}Cr, ^{60}Co, ^{54}Mn, ^{55}Fe and ^{59}Fe are examples of this type, which have long half-lives and cause problems by gamma emission. “Hot Particles” are small fragments of alloy, which become radioactive along with fission products. (NCRP, 1989)

Radionuclides accumulating in a reactor include fission products and activation products (those produced by neutron bombardment of impurities, such as those from corrosion, in the reactor). Fission products are produced in greater amounts compared to activation products; however, because of the nature of the reactor operation, activation products are predominant in nuclear wastes. Volatile fission products, such as iodine, are contained in the helium-filled gap.

3.2- Wastes from Nuclear Power Plants

The most popular type of reactor for commercial nuclear power plants is the light water reactor (LWR), including both boiling water reactors (BWR) and pressurized water reactors (PWR). In BWRs, coolant is directly converted to steam, which is then used to run a steam turbine. However, in PWRs, coolant is heated under high pressure, and is converted to steam in a second cycle and later used to run the turbine.

During normal performance of power plants, two types of wastes are generally obtained: liquid waste and gaseous waste. Both liquid and gaseous wastes consist of either fission products or induced activity nuclides in the primary coolant. Table 3-1 represents the initial activity in the core of a 3200 MWt nuclear Reactor. The composition of induced activity nuclides depends on the construction material of the core, reactor and other instrumentation that come into direct contact with water.

Table 3-1: Initial Activity of Radionuclides in the Core of a 3200 MWt Nuclear Reactor
From US Nuclear Regulatory Agency (1975)

Radionuclide	Amount (Ci)	Half-life
^{58}Co	0.0078	71
^{60}Co	0.0029	1,920
^{85}Kr	0.0056	3,950
^{85m}Kr	0.24	0.183
^{87}Kr	0.47	0.0528
^{88}Kr	0.68	0.117
^{86}Rb	0.00026	18.7
^{89}Sr	0.94	52.1
^{90}Sr	0.037	11,030
^{91}Sr	1.1	0.403
^{90}Y	0.039	2.67
^{91}Y	1.2	59
^{95}Zr	1.5	65.2
^{97}Zr	1.5	0.71
^{95}Nb	1.5	35
^{99}Mo	1.6	2.8
^{99m}Tc	1.4	0.25
^{103}Ru	1.1	39.5
^{105}Ru	0.72	0.185
^{106}Ru	0.25	366
^{105}Rh	0.49	1.5
^{127}Te	0.059	0.391
^{127m}Te	0.011	109
^{129}Te	0.31	0.048
^{129m}Te	0.053	0.34
^{131m}Te	0.13	1.25
^{132}Te	1.2	3.25
^{127}Sb	0.061	3.88
^{129}Sb	0.33	0.179
^{131}I	0.85	8.05
^{132}I	1.2	0.0958
^{133}I	1.7	0.875
^{134}I	1.9	0.0366
^{135}I	1.5	0.28
^{133}Xe	1.7	5.28
^{135}Xe	0.34	0.384
^{134}Cs	0.075	750
^{136}Cs	0.03	13
^{137}Cs	0.047	11,000
^{140}Ba	1.6	12.8

Table 3-1: Initial Activity of Radionuclides in the Core of a 3200 MWt Nuclear Reactor From US Nuclear Regulatory Agency (1975)

^{140}La	1.6	1.67
^{141}Ce	1.5	32.3
^{143}Ce	1.3	1.38
^{144}Ce	0.85	284
^{143}Pr	1.3	13.7
^{147}Nd	0.6	11.1
^{239}Np	16.4	2.35
^{238}Pu	0.00057	32,500
^{239}Pu	0.00021	8.9 x 10^6
^{240}Pu	0.00021	2.4 x 10^6
^{241}Pu	0.034	5,350
^{241}Am	0.00017	1.5 x 10^5
^{242}Cm	0.005	163
^{244}Cm	0.00023	6,630

Tritium is produced in large amounts in light water reactors. However, it is a low energy beta emitter and enters the environment in form of water, does not concentrate in biological systems and has a rapid turn over rate, which all together makes it not so dangerous. Other nuclides can enter the coolant either through cladding failure, which is dominated by more soluble fission products such as Cs and I, or diffusivity from the cladding. Coolant water can be purified by a combination of filtration and ion exchange processes.

Leakage of coolant from pumps, valves and flanges may also occur and introduce nuclides to wastewater. Other activities in the power plants including laundry and laboratory equipment washing also introduce nuclides to wastewater.

In pressurized water reactors, primary coolant may leak to the atmosphere. Also there can be some leakage from the heat exchanger, which can introduce the primary coolant to the second loop and consequently, the release of radioactive gases to the atmosphere without treatment, through boiler blow down. In boiling water reactors, fission products and radioactive noble gaseous may enter atmosphere through air leakage from condenser, which operates under vacuum. Several types of treatments can control these gaseous emissions.

4. RADIONUCLIDES FROM WEAPONS TESTS

The primary use of nuclear energy after its invention was the production of nuclear weapons. Nuclear science developed rapidly during World War II under war pressure. The first nuclear bomb was produced in US and the first atmospheric bomb test was conducted in 1945 by US. The United States, the former Soviet Union, U.K, France, China and some other countries continued these tests. Atmospheric nuclear weapon tests introduced large amounts of nuclides to the environment. The basics of nuclides release to the environment by the weapons test are briefly discussed in the following section.

Debris from nuclear weapons explosions depend on burst height and explosive yield, which dictates the height of the fireball. The conditions can be categorized in three groups:

1) Large particles which are highly radioactive and deposit within the first few hours after detonation,
2) Particles, which are small enough to act like aerosols and reach the troposphere. These do not deposit within the first day and their average lifetime in the troposphere is about 1 month. They mostly disperse within the latitude of release. Weapons in 100 kT range produce this type of debris. However, radionuclides associated with this group are usually

short-lived nuclides. As a result, when they deposit on soils and crops, many will decay before entering the food chain and human body,

3) Particles enter the stratosphere when produced by weapons larger than 500kT. These particles cause worldwide contamination.

Fission products released from weapon tests up to 1990 are estimated to be 2 x 10^{20} Bq, which is mostly spread through the same latitude of release, in the northern hemisphere. Weapons tests were also performed underground, on the surface and in the sea. Atmospheric tests began to raise concerns in 1950s. In 1963 the three nuclear powers, US, Russia and UK agreed to stop atmospheric tests, however; they continued performing underground tests and were committed to prevent venting from underground tests to the atmosphere.

Radioactive debris from nuclear detonations can be classified as: 1) Principal fission products, which include 200 isotopes of 35 elements, and 2) Induced nuclides, which are produced by neutron interaction with non-radioactive elements in the bomb materials, atmosphere, seawater or soil. Libby (1958) estimated that 3.2 x 10^{23} atoms of ^{14}C are produced from each kT of nuclear explosion due to this second mechanism.

It should be noted that some nuclides are not produced immediately after the explosion and they are decay products of primary fission products, such as ^{90}Sr, which is the second decay product of ^{90}K.

The height of explosion affects the type and amount of fallout. Near surface explosions produce large glassy particles which settle within the few hours; however, tests above the ground in the atmosphere produce steam like particles, which suspended in the atmosphere for a considerable amount of time and do not grow more than 3 micrometers in diameter, settling slowly.

There are several factors affecting fallout, including:

1) Explosive yield of the device,
2) Height of the explosion above the surface,
3) Meteorological conditions and
4) Design of the device

However, all the debris from a <500 kT bomb is deposited within the first two months at the same latitude following release to the atmosphere. Troposphere fallout is less responsible for long-lived nuclides, however; it is more responsible for short-lived nuclides such as ^{131}I. There is not much stratospheric contamination from kiloton explosion and mostly megaton explosions are responsible for stratospheric contamination. Fallout in the immediate region of an explosion is approximately 80 percent for land explosions, 20 percent for surface water explosions and 10 percent for atmospheric explosions.

The short-term biological effect from fallout depends on the rate at which the total dose was delivered and whether there was any food contamination. Fallout may result in fresh food contamination (mostly by ^{131}I) and contamination of leafy vegetables and water.

Table 4-1 shows the yields of principal nuclides per megaton of fission. Also Table 4-2 represents radionuclides produced and dispersed globally in atmospheric testing. Some of the more important nuclides from fallout will be discussed in the following section.

Table 4-1: Approximate Yield of the Principal Radionuclides per Megaton of Fission
From Klement (1965) and Knapp (1963)

Radionuclide	Half-life	Amount (MCi)
^{89}Sr	53 d	20
^{90}Sr	28 yr	0.1
^{95}Zr	65 d	25
^{103}Ru	40 d	18.5
^{106}Ru	1 yr	0.29
^{131}I	8 d	125
^{137}Cs	30 yr	0.16
^{131}Ce	1 yr	39
^{144}Ce	33 d	3.7

Table 4-2: Radionuclides Produced and Globally Dispersed in Atmospheric Nuclear Testing
From UNSCEAR 2000- Continued

Radionuclide	Half-life	Fission Yield	Normalized	Global Release
^{3}H	12.33 yr		740	186000
^{14}C	5730Yr		0.85	213
^{54}Mn	312.3 d		15.9	3980
^{55}Fe	2.73 yr		6.1	1530
^{89}Sr	50.53 d	3.17	730	117000
^{90}Sr	28.78 yr	3.5	3.88	622
^{91}Y	58.51 d	3.76	748	120000
^{95}Zr	64.04 d	5.07	921	148000
^{103}Ru	39.26 d	5.2	1540	247000
^{106}Ru	373.6 d	2.44	76	12200

Table 4-2: Radionuclides Produced and Globally Dispersed in Atmospheric Nuclear Testing
From UNSCEAR 2000

^{125}Sb	2.76 yr	0.4	4.62	741
^{131}I	8.02 d	2.9	4210	675000
^{140}Ba	12.75 d	5.18	4730	759000
^{141}Ce	32.5 d	4.58	1640	263000
^{144}Ce	284.9 d	4.69	191	30700
^{137}Cs	30.07 yr	5.57	5.9	948
^{239}Pu	24110			6.52
^{240}Pu	6563 yr			4.35
^{241}Pu	14.35 yr			142

4.1- ^{89,90}Sr

Strontium is one of the important nuclides from the fallout because of its long half-life of 28 years. It was estimated by the US Department of Energy that all ^{90}Sr released to the atmosphere up to 1963 was from weapon tests, deposited on the earth surface by 1970. It can be transferred to plants either by direct uptake from soil or by deposition on leaves; however, the most important source of this nuclide is milk. Uptake of this nuclide depends on the way food is processed and eating habits. It can be up taken as a substitute for Ca. Therefore, it is mostly found in human bone and its deposition on bones depends on variation of dietary intake over time and change in the rate of calcium accretion and turnover.

^{89}Sr is completely similar to ^{90}Sr except that it has much shorter half-life of 53 days and deposition on leaves is its most important source of contamination.

4.2- ^{137}Cs

It has a long half-life of 30 years and produced more abundantly than ^{90}Sr. Morisawa, et al (2005) reported that the fallout of ^{137}Cs was higher than ^{90}Sr in Japan after Chernobyl. ^{137}C moves slowly in soil and unless physically distributed, remains in the first 15-30cm of undistributed soil depending on soil type. Deposition of ^{137}Cs from fallout peaked in US in 1960's and an exception was a peak in 1986 due to fallout from the Chernobyl accident, which was insignificant in comparison to the deposition from weapon tests (Morisawa, et al 2005). Average ^{137}Cs concentration in northeastern US soil is reported to be 0.1-1 pCi/g. (The Health Physics Society 39th Annual Meeting Student III – Environmental and Radon Session June 28, 1994)

Aoyama et al, (2005) estimated total global ^{137}Cs fallout of 765±79 PBq for the northern hemisphere, which is 1.4 times higher than that the 545 PBq estimate in the UNSCEAR reports. ^{137}Cs bonds strongly with surface soil and as a result is not transferred to vegetation metabolically. Its biological half-life in human body is a function of age and sex.

4.3- ^{14}C

As mentioned before, ^{14}C is an induced nuclide produced by cosmic neutrons and the detonation of nuclear weapons. It has a long half-life of 5715 years and persists in the environment for many thousands of years. NCRP (1985) estimated the activity of stratospheric air to be 360 PBq. By the end of 1967, ^{14}C level reached 60 percent more than its natural background. NCRP (1985) estimated the dose to humans in the US to be from ^{14}C from fallout to be 0.96 mrem/year and 0.37 mrem/year in 1965 and 1984 respectively.

4.4- Pu

Plutonium can be released by nuclear detonations either as unfissioned or by neutron irradiation of ^{238}U. An alpha spectrometer cannot distinguish between ^{239}Pu and ^{240}Pu, so the amount of these two nuclides all-together is reported as measured plutonium. It is estimated that 12 PBq of plutonium has been released prior to 1963 globally by mostly megaton detonations. Lungs, osteocytes and livers will receive the highest doses from inhaled ^{239}Pu and ^{241}Am, which is an alpha emitter produced from ^{241}Pu.

4.5- ^{131}I

This nuclide is the most important one among iodine isotopes released by nuclear detonations. It has been estimated that 4600 PBq ^{131}I is released per megaton of fission and 10^6 PBq of this isotope were released through 1980 from nuclear weapon tests, mostly in megaton range. Most of the ^{131}I released from megaton detonations was transferred to stratosphere and decayed before reaching troposphere and depositing on earth. However, small amounts from megaton bombs and most of the ^{131}I from kiloton bombs will remain in the troposphere.

This nuclide has a short half-life of 8.2 days. Cattle can ingest it shortly after deposition and as a result ^{131}I will be present in fresh milk and other fresh dairy products, and consequently enters human body by consuming contaminated products. The thyroid gland receives highest dose from radioiodine.

5. RADIONUCLIDES FROM ACCIDENTS

Several accidents have happened since the start of the nuclear industry, each of which has its own degree of importance based on the psychological, environmental or economical impacts. The most recent nuclear facility accident was the 2011 Fukushima Daiichi power plant accident in Japan, following the tsunami. Although there are some immediately observable environmental impacts of this accident, many aspects of the accident will only become clear as time goes by. Like all the other accidents that have happened so far, a better understanding of the releases and their environmental impacts will be developed over time. The 1986 Chernobyl accident is the most disastrous one so far in terms of the amount of nuclides released. In the following section some of the well known nuclear accidents are discussed briefly.

5.1- Windscale (UK, 1957)

The Windscale facility consisted of two air-cooled, graphite moderated natural uranium reactors, used primarily for plutonium production. The accident was caused by the release of energy stored in crystalline structure of graphite carbon (known as Weigner's fever), which caused a rise in core temperature and subsequent combustion. This caused the release of a great amount of nuclides, primarily isotopes of volatile elements and noble gases by 125m tall ventilation stacks. However, stack filtration prevented release of some nuclides. The nuclide of most concern in this accident was ^{131}I, which was observed in cows' milk on the afternoon of first day. Chamberlain and Dunster (1958) studied the form of the released activity and concluded that most of the released radioiodine was in gaseous form or attached to very small particulate materials. Eisenbud and Gesell[1], 1997, Clarke[2], 1974 and Garland and Wakeford[3], 2007, studied the amount of release inventory for ^{131}I to be 599-740, 600, 900-3700 Tbq respectively. Other nuclides estimated are represented in Table 5-1.

Table 5-1 Estimation of the Amount of Radionuclides Released in Windscale Accident From Eisenbud and Gesell[1] (1997), Clarke[2] (1974) and Garland and Wakeford[3] (2007)

Radionuclide	Estimated amount released in TBq
^{137}Cs	22.2-45.9[1], 45.5[2], 90-350[3]
^{89}Sr	2.9-5.1[1], 5.1[2], 6-100[3]
^{90}Sr	0.2-0.2[1], 0.22[2], 0.18-3.1[3]

Table 5-2 Types and Amount of Radionuclides Released During the First Days After the Chernobyl Accident, UNSCAER 2000 (Last four column calculated by the author)

Radionuclide	Half-life	Release (Total EBq)	Initial %	Amount after 100d	% After 100d	Amount After 30y	% After 30y
^{85}Kr	10.73 y	0.033	0.75	0.0324	9.8	0.0048	17.6
^{89}Sr	50.5 d	0.094	2.14	0.0238	7.19	~0	
^{90}Sr	28.6 y	0.0081	0.18	0.008	2.42	0.0039	14.3
^{95}Zr	64 d	0.16	3.64	0.054	16.32	~0	
^{103}Ru	39.4 d	0.14	3.19	0.024	7.26	~0	
^{106}Ru	368 d	0.059	1.34	0.049	14.81	~0	
^{131}I	8.04 d	0.67	15.27	0.00012	0.04	~0	
^{133}Xe	5.24 d	1.7	38.74	$3.06\ \mathrm{x}10^{-6}$	~0	~0	
^{134}Cs	2.07 y	0.019	0.43	0.017	5.14	$8.2\mathrm{x}10^{-7}$	0.003
^{136}Cs	13.2 d					~0	
^{137}Cs	30.2 y	0.037	0.84	0.0368	11.12	0.0185	67.8
^{140}Ba	12.8 d	0.28	6.38	$1.24\mathrm{x}10^{-3}$	0.37	~0	
^{141}Ce	32.5 d	0.13	2.96	0.0154	4.66	~0	
^{144}Ce	284 d	0.088	2	0.069	20.86	~0	
^{239}Np	2.36 d	0.97	22.1	~0	~0	~0	
^{238}Pu	87.7 y	$3\mathrm{x}\ 10^{-5}$	$6.84\mathrm{x}10^{-4}$	$2.99\mathrm{x}10^{-5}$	0.009	$2.37\mathrm{x}10^{-5}$	0.09
^{239}Pu	24100 y	$2.6\mathrm{x}\ 10^{-5}$	$5.92\mathrm{x}10^{-4}$	$2.6\mathrm{x}10^{-5}$	0.008	$2.6\mathrm{x}10^{-5}$	0.09
^{240}Pu	6570 y	$3.7\mathrm{x}\ 10^{-5}$	$8.43\mathrm{x}10^{-4}$	$3.7\mathrm{x}10^{-5}$	0.011	$3.7\mathrm{x}10^{-5}$	0.13
^{241}Pu	14.4 y						

5.2- Three Mile Island (US, 1979)

TMI Unit 2 accident was the first accident in a civilian power reactor, and resulted in a major down turn in nuclear power plant development. This accident was caused as a result of loosing reactor coolant. 50 percent of the core melted during the accident; however, fission products mainly consisting of Xe, Kr and about 1 TBq of ^{131}I remained in the building and radioactive exposure to plant workers and general public was minor. Waste management, specially handling contaminated water, was the main post-accident issue. Hundreds of gallons of contaminated water with concentration of about 1 GBq/ m^3, mostly consist of ^{137}Cs, accumulated in the reactor-building sump. Ion exchange process where designed to decontaminate these water and the decontamination was sufficiently effective that the only nuclide remained in the water was tritium. The levels of this nuclide were low enough to allow the water release in nearby streams, but were not acceptable by general public. Eventually, after 10 years of holding the contaminated water in tanks, large, expensive heat exchangers where built to evaporate it.

5.3- Chernobyl (Former USSR, 1986)

So far, this accident is the worst one among civilian nuclear power reactors in terms of the amount of radioisotopes released and environmental impacts, considering that not much knowledge is available about Fukushima. It happened while performing some tests on Reactor 3 after operating at full-power level of 3200MWt, and caused a steam explosion. Around 200 different radionuclides were released by this accident, which later caused global contamination by fallout. The reactor fuel was 610 days old and reactor core contained the residue of $5.6x10^{27}$ fissions. Four EBq ($1E = 10^{18}$) fission products approximately were released in the European part of former Soviet Union. The total amount of radionuclides released to the environment was about 11 EBq. About 2 EBq of radionuclides were released to the atmosphere during the first days after the accident. The ^{137}Cs realize was approximately 100 PBq. Also over 185 PBq (1P

= 10^{15}) of ^{131}I, which caused thyroidal doses among children in the surrounding general population as high as several grays, were released.

As mentioned, this accident caused worldwide contamination and several studies concentrated on impacts of it all around the world, special in the northern hemisphere. McAulay and Moran (1988) reported the total ^{137}Cs deposition of approximately 2.6x10^{14} Bq over Ireland, which is between 0.4-0.5% of the worldwide ^{137}Cs deposition. The average ^{137}Cs soil contamination over Ireland averaged 3.2 kBq/m^2 over Ireland.

Table 5-2 shows the type and amount of nuclides released during the first days after the accident. (UNSCAER, 2000) The last four columns of this table were by calculated by the author by adding the radioactive decay of each nuclide using its half-life.

Radioactive materials were released as gas and dust particles and were first transported by wind. Two types of radioactive particles were released by accident. The first group including larger airborne particulates were composed of finely dispersed reactor fuel, and the second group, smaller particles, were nuclides formed during the graphite fuel fire (such as I, Te, Cs, and a lesser amount of Sr and Ru). The second group was distributed over a greater distance. After the decay of short-lived nuclides and deposition of long-lived ones, such and Cs and Sr, by fallout during past thirty years, the most important mechanism of radionuclides transport is from soluble nuclides by contaminated water, rather than dispersion by wind. This pattern of evolving mechanism of dispersion is likely to be true for other accidents or releases.

5.4- Fukushima Diichi (Japan, 2011)

An earthquake of magnitude 9.0 in Great East Japan primarily led to this accident. The earthquake on 11 March 2011 was followed by a tsunami, which eventually caused damage to power supply system and seawater pumps causing an interruption of cooling to reactors of unit 1,2 and 3. This interruption caused the accident in units 1, 2 and 3 in the first day and unit 4 in the fifth day. This accident, like all the others, caused release of radioactive material to both the immediate environment and worldwide. ^{131}I, ^{137}Cs, ^{134}Cs, ^{239}Pu, ^{241}Pu and possibly some ^{35}S were easily observed. From 12 March to 6 April 2011, an estimated 150 PBq ^{131}I and 13 PBq ^{137}Cs were released to the atmosphere. (Chino et al, 2011) suggested a higher value of 50 PBq for ^{137}Cs was released to the northern hemisphere. 70-80 percent of radionuclides released were deposited over the western North Pacific Ocean. On March 23, oceanic contamination in seawater 30km offshore of the plant was up to 77 Bq/L for ^{131}I and 24 Bq/L for ^{137}Cs. By the end of May, 4 PBq of ^{137}Cs was released by direct discharge to seawater. By June 2011, offshore ^{137}Cs readings were below detection limits. (Yoshida and Kanda, 2011) has suggested that the rapid decline in Cs was due to precipitation and sedimentation.

Deposition of ^{137}Cs caused high contamination of nearby soils. In the case of deposition, this nuclide remains in the first few centimeters of soil. In nearly half of the 20 km exclusion zone around nuclear power plant, ^{137}Cs deposition exceeded 600,000 Bq/m^2 and in highly contaminated areas deposition exceeded 3,000,000 Bq/m^2.

This accident also caused worldwide contamination. The effect of this accident is only about the tenth of Chernobyl. Fission products, ^{131}I, ^{132}I, ^{132}Te, ^{134}Cs, ^{137}Cs, reached US in 17-18 March 2011 and were detected in milk, air and water samples. The highest detected activity values ranged from less than 1 mBq/m^3 to 31 mBq/m^3 for the particulate ^{131}I, and up to 96 m Bq/m^3 for the gaseous ^{131}I fraction. The maximum concentrations of ^{131}I measured over the United States were around 70.3 mBq/m^3 on March 21, 2011 in Anaheim, California and 103.6 mBq/m^3 on March 30, 2011 in Dutch Harbor, Alaska, while for most other part the activity remained typically low or non-detectable. (P. Thakur et al, 2011)

6. RADIONUCLIDES IN WATER SUPPLIES

Different types of nuclides have been observed in surface and ground waters. Before the start of the nuclear programs, most of these nuclides were the naturally occurring ones. These nuclides are primarily introduced to water due to its contact with rocks and minerals. It is also possible that in some areas, radioactive gases from molten magmas transfer some nuclides to water. This was the general trend before the start of the nuclear programs. However, large amount of nuclides can be introduced to water supplies by fallout. Fission products from nuclear tests and accidents have been distributed in rain and consequently find their way to water supplies. If these nuclides enter the human body, their effects may not become apparent for several generations.

6.1- Natural Radioactivity in Water

Radionuclides from three natural radioactive chains, which are present in rocks and minerals, find their way to surface and ground waters. Their concentration depends on factors such as half-life and solubility. ^{238}U, ^{226}Ra and its gaseous ^{222}Rn decay product, are the only nuclides from the Uranium series found in water (Love, 1951). Thorium occurs in nature in larger quantities than uranium. According to Rankama and Sahama, the average thorium-uranium ratio for igneous rocks is about 3. However, very few data are available on the thorium content of water and this occurs because of its relative low solubility.

Radionuclides can be found in seawater. Karlik found that the uranium content in seawater ranges from 1.1 to 1.7 x 10^{-6} gr/l, depending on the geographic source of water and to a certain extent to the depth at which the samples were obtained. Radium content of seawater; however, is not proportional to its uranium content and measurements show values far below those calculated assuming uranium-radium equilibrium. Evans (1938) and Petterson (1939) measured the average content on radium in seawater to be 0.08x10^{-12} gr/l. This lesser amount of radium compared to the amount in equilibrium with uranium may occur because of uptake by marine animals, chemical precipitation or physical processes such as adsorption and ion exchange with sediments. Foyn and Rona measured the thorium content of seawater and the maximum concentration of 0.5 x 10^{-6} gr/l was generally observed.

High concentrations of nuclides, specifically, radium, have been observed in spring waters. Natural spas were well known for their mysterious power of healing diseases long before their radionuclide content was known. The amount of radium in spring waters ranges over wide limits. In the United States, the maximum published value for spring water is for Curie Spring near Boulder, Colorado, which is reported by Fitch to contain 267,800 x 10^{-12} gr/l radium. However, values as high as 703,000 x 10^{-12} gr/l have been reported for hot springs in Japan. These high amounts of radium are introduced to these spring waters due to their contact with igneous rocks or uranium deposits.

The radioactivity of ground water has been studied by Judson (1955) in the Colorado plateau. According to this study, ground waters in contact with mineralized deposits (100-460 ppb) were more radioactive than those in contact with non-mineralized deposits (2-9 ppb).

Drinking water taken from drilled wells may contain high concentrations of natural radionuclides. The most significant radionuclides typically found in drinking water, with respect to doses, are isotopes of uranium, radium, polonium, lead and ^{222}Rn. According to the USEPA the maximum contamination level for radium, including both ^{226}Ra and ^{228}Ra, is set to 0.19 Bq/l and for gross alpha (including radium but excluding uranium and radon) to 0.56 Bq/l.

The concentration of nuclides in drinking water is well controlled to be below these limits. However, some elevated levels of radionuclides have been observed. Outola (2004) reported high levels of ^{210}Po in drinking water samples in two different areas of Maryland. The ^{210}Po activity in drinking water samples of Charles county were reported to be 1700 ± 100 mBq/l and the gross alpha activity is solely due to ^{210}Po presented in water. The removal of polonium is not well achieved by ion exchange or activated carbon units. However revers osmosis is efficient in polonium removal.

Also elevated levels of nuclides were observed in Baltimore County (Outola, 2004) with noticeable amount of radium. Water softener processes can remove up to 90% of radium. However, the best method for removing this nuclide from drinking water is reported by Zapecza (1986) to be a combination of ion exchange using strong acid cation resins and reverse osmosis which can remove up to 99% of radium. However, this method may not be effective in removing uranium, because it is normally present in water in anionic complexes.

Nuclear accidents can introduce some radionuclides to water supplies. These nuclides may be introduced directly from fallout or by washing of radionuclides to surface waters after rainfall. Imhoff (1988) report no critical concentrations of radionuclides in potable water from Ruhr basin, Germany after Chernobyl accident. Some high levels of ^{131}I were observed. The highest concentration of ^{131}I observed in Ruhr River was 22.6 Bq/l; however, this nuclide was not of high concern for the evaluation of potential harm to drinking water because of its short half-life. The amount of radionuclides released to this basin due to Chernobyl fallouts were well below what introduced during nuclear tests of 1962.

7. RADIONUCLIDES IN WASTEWATER TREATMENT PLANTS

Radioisotopes, which are found in wastewater treatment plants, can come from different sources. Naturally occurring radionuclides were observed in treatment facilities even before the start of the utilization of radionuclides by humans in industrial, medical and other activities. After the beginning of nuclear activities, radionuclides were introduced to wastewaters from laboratories performing studies on radionuclides by direct discharge of nuclides or by other indirect activities such as discharging contaminated laundry wastewater.

Medical facilities are large contributors of certain nuclides to treatment plants. Some industrial facilities like those engaged in radioactive waste processing and nuclear power plants also introduce radioisotopes to treatment plants. Accidents at some nuclear power plants have released high doses of nuclides to atmosphere, water streams and eventually wastewater plants. After the start of atmospheric weapon tests, high doses of fission products were introduced to wastewater by fallout or washing of nuclides from contaminated surfaces.

After entering the treatment facilities, radionuclides will fractionate between liquid and solids phases based on their physical and chemical characteristics and the type of treatment process. Some of them may even decay before being discharged from the treatment plant due to their short half-lives.

In the following section, the presence of radionuclides in wastewater treatment effluent and sludge are discussed.

Several radionuclides from different sources with different concentrations have been observed in treatment plants. The goal is to keep these concentrations under the assigned limits for each of these radionuclides. Limits are set based on the characteristics of a nuclide, including its half-life and decay type and energy. Some nuclides like ^{99m}Tc, which has medical usage and has been observed in several treatment plants close to the medical facilities. ^{90m}Tc has a short half-life of only 6 hours and would likely spend sufficient time before leaving the treatment plant to decay and reach low activity. These types of radionuclides are of less concern, unless their concentrations is so high to cause health problems for treatment plant operators and staff.

Unfortunately not all radionuclides in wastewater treatment plants have short half-lives of few hours. ^{131}I, which is medically used and also a fission product, is a common radionuclide in treatment plants close to medical facilities and industries, which produce fission products such as nuclear power plants. ^{131}I is a beta emitter with half-life of 8 days. Its half-life is not as long as nuclides such as ^{137}Cs, which provides the opportunity for decay before leaving the treatment plant, whether as attached to sludge or in the effluent. Because of its frequent use in medicine and industry, period or sporadic releases of this nuclide occur. Its short half-life and therefore high rate of decay can cause highly contaminated discharges to the environment.

Other types of nuclides with much longer half-lives such as Cs and Sr have also been observed in treatment plants. Different isotopes of cesium have been observed in wastewater treatment influents. ^{134}Cs and ^{137}Cs are of concern because of their long half-lives. ^{137}C, which is a beta emitter with 30 years half-life, may appear in treatment plants from different sources. This nuclide has medical uses, but it is mostly released to environment and sewage systems as a fission product. Nuclides like ^{137}Cs do not have the chance to decay during the treatment process and will leave the treatment plant weather as attached to sludge or soluble in the effluent.

Several radionuclides observed in treatment plants and their concentrations in different parts of the plant are presented in Table 7-1. These observations were collected from the available literature, worldwide.

Table 7-2 is reduction from Table 7-1 and some other data to show the fate of the various nuclides in a treatment plant. Only those cases where there is sufficient information have been analyzed and included in Table 7-2. In many cases it was necessary to make assumption or calculate intermediate variables. The goal of this analysis was to determine the likely fate of the various nuclides in treatment plants, based upon the various and sometimes-accidental observations. An important finding is to determine the partitioning of the various nuclides between the liquid and solid phases, since the two phases are discharged differently from treatment plants.

Among different contributors of radionuclides to the environment and wastewater system specifically, medical facilities are the most regular and consistent ones and nuclear accidents are the least frequent but disastrous ones in terms of the amount of nuclides released. In the following section radionuclides released to treatment plants by medical facilities as one of permanent sources will be reviewed in detail.

Table 7-1. Radionuclides observed in different wastewater treatment plants and their partition between liquid and solid phases

Nuclide	Half-life	Liquid Effluent	Primary Sludge	Digested Sludge	Reference	Location
^{131}I	8.06d		1% of total		Martin JE	
		ND		45 pCi/kg(1.665 Bq/kg)	Durham	Hamilton plant,canada
		ND		185 pCi/kg(6.845 Bq/kg)	Durham	Dundas plant,Canada
				<10-1600 Bq/kg	Imhof	Duisburg-Kaberfeld
				<10-7180 Bq/kg	Imhof	Arnsberg
				<10-3370 Bq/kg	Imhof	Hagen
				<10-2570 Bq/kg	Imhof	Warstein
				<10-2520 Bq/kg	Imhof	Essen-Brugaltendorf
				<10-1370 Bq/kg	Imhof	Meinerzhagen
				1034 Bq/kg	Imhof	Hattingen
				420 Bq/kg	Imhof	Witten
				<10-3230 Bq/kg	Imhof	Ruthen
^{137}Cs	30.2 yr		1.7 pCi/gr(62.9 Bq/kg)	3.0 pCi/gr(111 Bq/kg)	Folsom	Hyperion
		0.025pCi/l		210 pCi/kg(7.77 Bq/kg)	Durham	Hamilton plant,canada
		ND		100 pCi/kg(3.7 Bq/kg)	Durham	Dundas plant,Canada
				160-2700 Bq/kg	imhof	Duisburg-Kaberfeld
				900-10,380 Bq/kg	imhof	Arnsberg
				1190-3830Bq/kg	imhof	Hagen

Table 7-1. Radionuclides observed in different wastewater treatment plants
and their partition between liquid and solid phases- Continued

Nuclide	Half-life	Liquid Effluent	Primary Sludge	Digested Sludge	Reference	Location
				1300-5620 Bq/kg	imhof	Warstein
^{131}I	8.06d			800-6990 Bq/kg	imhof	Essen-Brugaltendorf
				1710-6170 Bq/kg	imhof	Meinerzhagen
				4850 Bq/kg	imhof	Hattingen
				5900 Bq/kg	imhof	Witten
				3930-9150 Bq/kg	imhof	Ruthen
			0.2pCi/gr(avg) (9.62 Bq/kg)	0.38pCi/gr(avg) (14.06 Bq/kg)	Folsom	Hyperion
				0.6-56 Bq/kg	Erlandsson	Borgby,Sweden
				<1-2.4 Bq/kg	Erlandsson	Borgby,Sweden
			3.3x10-3gr/gr	6.3x10^{-3}(gr/gr)	Folsom	Hyperion
			-	-	Folsom	Hyperion
		ND		1024 pCi/kg (37.88 Bq/kg)	Durham	Hamilton plant,canada
		ND		634 pCi/kg (23.46 Bq/kg)	Durham	Dundas plant,Canada
^{228}Th	1.9yr	ND		236 pCi/kg (8.73 Bq/kg)	Durham	Hamilton plant,canada
		0.02pCi/l (0.0007 Bq/l)		284 pCi/kg (10.51 Bq/kg)	Durham	Dundas plant,Canada
^{232}Th			2.8x10-6gr/gr	5.3x10-6gr/gr	Folsom	Hyperion

Table 7-1. Radionuclides observed in different wastewater treatment plants and their partition between liquid and solid phases

Nuclide	Half-life	Liquid Effluent	Primary Sludge	Digested Sludge	Reference	Location
^{51}Cr	27.8d	1.24pCi/l (0.0459 Bq/l)		2705 pCi/kg (100 Bq/kg)	Durham	Hamilton plant,canada
		ND		ND	Durham	Dundas plant,Canada
^{106}Ru	1yr	ND		951 pCi/kg (35.19 Bq/kg)	Durham	Hamilton plant,canada
^{103}Ru	39d			5.4-18.1 Bq/kg	Erlandsson	Borgby, Sweden
^{144}Ce	284d	ND		1248 pCi/kg (46.18 Bq/kg)	Durham	Hamilton plant,canada
		ND		676 pCi/kg (25.012 Bq/kg)	Durham	Dundas plant,Canada
^{141}Ce	32.5d	ND		145 pCi/kg (5.36 Bq/kg)	Durham	Hamilton plant,canada
		ND		ND		Dundas plant,Canada
				4.7-9.9 Bq/kg	Erlandsson	Borgeby,Sweden
^{95}Zr	64d	ND		254 pCi/kg (9.40 Bq/kg)	Durham	Hamilton plant,canada
				25-53 Bq/kg	Erlandsson	Borgeby,Sweden
^{95}Nb	35.1d	ND		364 pCi/kg (13.47 Bq/kg)	Durham	Hamilton plant,canada
^{7}Be	53.6d	0.23pCi/l		2227 pCi/kg (82.40 Bq/kg)	Durham	Hamilton plant,canada
		ND		827 pCi/kg (30.60 Bq/kg)	Durham	Dundas plant,Canada
				30-61 Bq/kg	Erlandsson	Borgeby,Sweden
^{24}Mn				0.9-5.2 Bq/kg	Erlandsson	Borgeby,Sweden

Table 7-2. Radionuclides in Wastewater treatment plants and
their partition between liquid and solid phases

		Influent (total)(GBq)	Effluent (GBq)	Sludge (GBq)	%Effluent	%Sludge	Ref
<1-20% in Sludge	^{131}I	0.296	0.2351	0.0609	79.42	20.58	Barquero,R
	Hospital	3.219	3.1883	0.0306	99.05	0.95	Erlandsson,B
	Hospital	1.7427	1.7172	0.0255	98.54	1.46	Erlandsson,B
	Bomb Test	8.288	8.2237	0.0642	99.22	0.78	Erlandsson,B
		(Sum of effluent water and sludge)	0.0672	0.0076	89.79	10.21	Erlandsson,B
			0.0526	0.0113	82.32	17.68	Prichard
			0.0944	0.0069	93.18	6.82	Prichard
28-38% in Sludge	^{51}Cr	(Sum of effluent water and sludge)	0.0064	0.0040	61.13	38.87	Erlandsson,B
			0.0139	0.0061	69.43	30.57	Erlandsson,B
			0.0204	0.0080	71.80	28.20	Erlandsson,B
	Mean value				67.46	32.54	
34-71% in Sludge	^{137}Cs	(Sum of effluent water and sludge)	0.00294	0.0019	61.25	38.75	Erlandsson,B
			0.00245	0.0021	54.20	45.80	Erlandsson,B
			0.00116	0.0029	28.64	71.36	Erlandsson,B
			0.00203	0.0023	46.88	53.12	Erlandsson,B
			0.00301	0.0030	49.67	50.33	Erlandsson,B
			0.00536	0.0028	65.85	34.15	Erlandsson,B
	Mean value				51.08	48.92	

8. MEDICALLY USED RADIONUCLIDES IN WASTEWATER TREATMENT PLANTS

Radionuclides are used in medical facilities for radiotherapy, radio diagnoses and research. According to United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2000) more than 30 million diagnostic nuclear medicine procedures are carried out each year. Radionuclides such as ^{131}I are commonly used for radiotherapy purposes, and according to the same study by UNSCEAR, approximately 90% of current nuclear medical therapies use this radionuclide. These radionuclides can be reactor-produced or accelerator-produced. Table 8-1 presents the reactor-produced and accelerator-produced radionuclides with their specific medical application (from Adelstein, 1995).

Table 8-1. Radionuclides and their specific medical application

^{77}As	In Cancer Therapy
^{82}Br	In metabolic studies and studies of estrogen receptor content
^{47}Ca	In studies of cell function and bone formation of mammals and to produce ^{47}Sc
^{252}Cf	In brachytherapy for treatment of cervical cancer and potentially for treatment of gliomas
^{14}C	For medical research to trace metabolism of new drugs and other organic carbon-containing molecules
^{141}Ce	For research and development on lung densities
^{137}Cs	To treat cancer; to measure correct patient dosages of radiopharmaceuticals
^{51}Cr	To asses red blood cell survival studies
^{58}Co	To diagnose pernicious anemia
^{60}Co	To treat cancer and sterilize surgical instruments

Table 8-1. Radionuclides and their specific medical application-continued

^{64}Cu	As a clinical diagnostic agent for cancer and metabolic disorder
^{67}Cu	In cancer therapy and to label antibodies for cancer therapy
^{165}Dy	To treat rheumatoid arthritis
^{166}Dy	Decays to ^{166}Ho which is used in cancer therapy
^{253}Es	To radiolabel antibodies for cancer therapy
^{169}Er	To treat rheumatoid arthritis
^{255}Fm	To radiolabel antibodies for cancer therapy
^{186}Re	As a bone cancer therapeutic agent and to radiolabel various molecules as cancer therapeutic agents; also used to treat rheumatoid arthritis
^{188}Re	For treatment of medullary thyroid carcinoma and alleviation of pain in bone metastases
^{145}Sm	For treatment of ocular cancer
^{153}Sm	To radiolabel various molecules as cancer therapeutic agents and to alleviate bone cancer pain
^{47}Sc	In therapy of cancer
^{75}Se	In protein studies in life science research
^{111}Ag	In cancer therapy
^{85}Sr	To study bone formation and metabolism
^{89}Sr	To alleviate metastatic bone pain
^{90}Sr	Decays to ^{90}Y, which is used in cancer therapy
^{35}S	In studies of cell metabolism and kinetics, molecular biology, genetics Research, biochemistry, microbiology, enzymology, and as a starter to make many basic chemicals and research products
^{99m}Tc	The most widely used radiopharmaceutical in nuclear medicine imaging
^{123m}Te	For research and development on lung densities and calibrating; also used in cardiology
^{117m}Ti	For palliative treatment of bone cancer
^{3}H	To make tritiated water, which is used as a starter for thousands of different research products and basic chemicals, and for life science and drug metabolism studies to ensure the safety of potential new drugs
^{188}W	In nuclear medicine for lung ventilation and perfusion studies
^{90}Y	To radiolabel various molecules as cancer therapeutic agents
^{159}Gd	In cancer therapy
^{199}Au	In cancer therapy and to treat rheumatoid arthritis
^{166}Ho	In cancer therapy and to treat rheumatoid arthritis
^{125}I	As a potential cancer therapeutic agent and for basic biomedical research

Table 8-1. Radionuclides and their specific medical application-continued

^{129}I	To check radioactivity counters in in vitro diagnostic testing
^{131}I	To diagnose and treat thyroid disorders including cancer for basic biomedical research
^{191}Ir	TO assess cardiac function especially in the pediatric population
^{192}Ir	In cancer therapy
^{177m}Lu	In cancer therapy and to label antibodies for cancer therapy
^{99}Mo	To produce 99mTc, the most commonly used radioisotope in clinical nuclear therapy
^{191}Os	Decays to 191mIr, which is used for cardiac studies
^{194}Os	Decays to 194Ir, which is used in cancer therapy
^{103}Pd	In the treatment of prostate cancer
^{32}P	In cancer treatment, cell metabolism and kinetics, molecular biology, genetics research, biochemistry, microbiology, enzymology, and as a starter to make many basic chemicals and research products
^{33}P	In cancer treatment, molecular biology and genetics research, and biochemical and enzymological studies
^{195m}Pt	In pharmacokinetic studies of antitumor agents

The half-lives of these nuclides range from less than a second to several years and the type of emission they cause by decay and the energetics of decay differ among them.

Several doses of nuclides can be used for each type of medical procedure. As an example, in case of the thyroid cancer treatment, approximately 4.14 GBq of ^{131}I is used during each treatment and in the case of hyperthyroidism, 414 MBq of this isotope is administrated for each treatment. (Barquero, 2008)

These nuclides, after remaining for a period of time in a patient's body, will be excreted by patient's urine and make their way to the wastewater treatment system. However, the amount of the radionuclides being discharged by urine and eventually treatment plants depends on the protocol used for the treatment of liquid waste and varies for inpatient and outpatients. Medical treatments received on an inpatient basis require the patient to stay in special rooms for approximately 4-7 days after receiving treatment and during this time, the urine is collected and sent to the storage tanks before discharging to sewage. This reduces the activity of radionuclides being released to wastewater collection systems due to decay of nuclides during storage period; however, this may cause higher doses to hospital staff who must handle the urine bag as compared to outpatient-based treatment in where the urine is directly discharged to the sewage system. (Erlandsson, 1977)

The increased number of nuclear medical treatments, since their development, has caused concerns regarding radionuclides discharged to wastewater treatment plants and their accumulation in sludges and receiving waters. Contaminated sludge can cause high exposure to treatment plant workers. Also, sludge from the wastewater treatment plants, with or without digestion, are often used as fertilizers and soil conditioners, which in case of contamination with radionuclides, may introduce these nuclides to food chain and human body.

Several studies have been devoted to both measuring and modeling the concentration of medically used nuclides in the sewage sludges and effluents in the US and other countries. Ruchhoft and Feitelberg (1951) studied the concentration of ^{131}I in sewage sludge and effluents of hospitals in six large cities of the US and emphasized several factors influencing these concentrations, such as decay of nuclides during treatment processes, which were neglected in preceding studies. According to their study, the portion of radioactive or any other material being adsorbed on the sludge is dependent on the character of the material adsorbed, the character of the sludge, the quantity of sludge, the period of contact between the supernatant and the sludge and temperature. As an example Ruchhoft (1949) showed that activated sludge has higher adsorption power than raw sludge and concentrations up to 2000 to 3000 ppm of sludge can adsorb 95% of low quantities of plutonium during 24-hour contact time. Their results for iodine removal were in agreement with preceding studies, which measured 0 to 3.7 % removal of this nuclide from supernatant by activated sludge.

A total resistance time of 38 days for ^{131}I before it will be discharged from the plant, which is nearly 4 times the half-life of this nuclide were estimated in this study. The decay period involved and the dilution of iodine in sewage before reaching to the treatment plant prevents hazardous doses to plant workers and high concentrations in receiving bodies.

In another study by Picharnd et al. (1981), the concentration of ^{131}I in sludges and effluent of a wastewater treatment plant in Texas with 50 MGD capacity, which served 10 medical centers and hospitals were measured by gamma-spectrometry. Also the partition of this nuclide between sludge and liquid effluent were studied. In this research, the highest specific activity was noted in the end product of the sludge treatment process. In gamma-spectrometry of sludge, the highest peak was related to Iodine and an activity of 37.5 pCi/ gr of sludge was obtained at the time of sample collection. Also some other peaks confirmed the presence of ^{99m}Tc and ^{95}Zr-^{95}Nb complexes. The concentration of Iodine in effluent was measured by concentrating this nuclide utilizing an ion exchange system and ^{131}I concentration of 111±2.1 pCi/lit of unconcentrated effluent were measured. At the time of this study, the ratio of Iodine in liquid effluent to that in dried sludge was 3.3. A total annual release of 1.6 Ci and 2.5 Ci for ^{131}I and ^{99m}Tc to this treatment plant was estimated respectively. The results for Iodine release from this study was in agreement with observation of Shapiro and Moeller (1978) that the amount of ^{131}I excreted by patients of a large medical treatment facility can exceed that of a large commercial power plant.

Although ^{131}I is one of the most important radioisotopes released from medical facilities to the environment, mainly because of its long half-life, other nuclides are observed in wastewater treatment facilities serving hospitals.

Miller et al. (1996) found ^{137}Cs and ^{60}Co in sewage sludge samples from Missouri, US. Chisaka et al. (1977, 1982) studied the behavior of radiopharmaceuticals from wastewater treatment plants in Tokyo, Japan, and found almost all the ^{67}Ga, ^{75}Se, and ^{137}Cs in activated sludge and the ^{131}I in discharged water. Nakamura et al. (2005) studied the concentrations of ^{99m}Tc, ^{123}I, ^{67}Ga and ^{201}Tl in the liquid effluent and the sewage sludge from Kurume Central Sewage Treatment Plant, Japan. The average activities of these nuclides in discharged water were 1.5% for ^{99m}Tc, 1.5% for ^{123}I, 4.3% for ^{67}Ga, and 0.41%

for ^{201}Tl of their average injected doses and the concentrations are about 2.5 x 10^{-5} of the legal concentration limits of discharged water in Japan.

Erlandsson (1977) has studied concentrations of radionuclides in sludge of a sewage treatment plant serving the borough of Malmo, Sweden. This treatment plant serves one hospital and almost all of the patients live in the borough. In this study, the ^{131}I concentration due to medical use varied between 0.034±0.01 and 0.124±0.02 nCi/kg. High concentration of nearly 0.4 pCi/kg is due to a bomb test in China during the time of study. Other nuclides such as ^{198}Au and ^{201}Tl were also observed, but concentrations of these were usually below detection limits. Finally he concluded that the ^{131}I concentration of the sludge due to medical use of the radionuclide is low and does not introduce any radiation protection problems for the persons working at the sewage treatment plan.

9. CONCLUSIONS

This thesis has reviewed the naturally occurring and anthropogenic radionuclides and their likely fate in wastewater treatment plants. The naturally occurring radionuclides, including the uranium and thorium decay series are always present and represent chronic sources, which are not likely to change as a result of an accident or spill. Anthropogenic radionuclides such as ^{137}Cs, ^{131}I, ^{90}Sr and are normally present in very low concentrations, but could be produced in high concentrations from an accident. Because of their moderate half-lives, they produce the most risk. Reclamation activities are therefore at episodic risk from these radionuclides, and treatment plants should develop contingency plans for these specific isotopes.

Based on their chemical and physical characteristics, they will have different fates in treatment plants. ^{137}Cs will likely partition onto sludges and biosolids that be discharged in the liquid effluents. Because of its 30.7 year half-life, storage to segregate the radionuclides is either not feasible or a very long-term project. ^{131}I will more likely remain in the liquid effluent than partition onto the sludge. Also its short half-life of 8 days means that it will have limited effects if widely distributed. There is concern for the treatment plant workers and through short-term exposure. The fate of Strontium is less certain but its 28.6 year half-life and accumulation in the bones makes it an important concern.

Few treatment technologies exist for treatment of radionuclides in wastewaters at low concentrations. Enhancing coagulation-precipitation in treatment plants, such as chemically assisted primary clarification, is one possible treatment improvement. Segregation of contaminated waters and using volume reduction by evaporation or reverse osmosis is also a possibility. The thesis identifies areas of research needed to develop short-term treatment technologies.

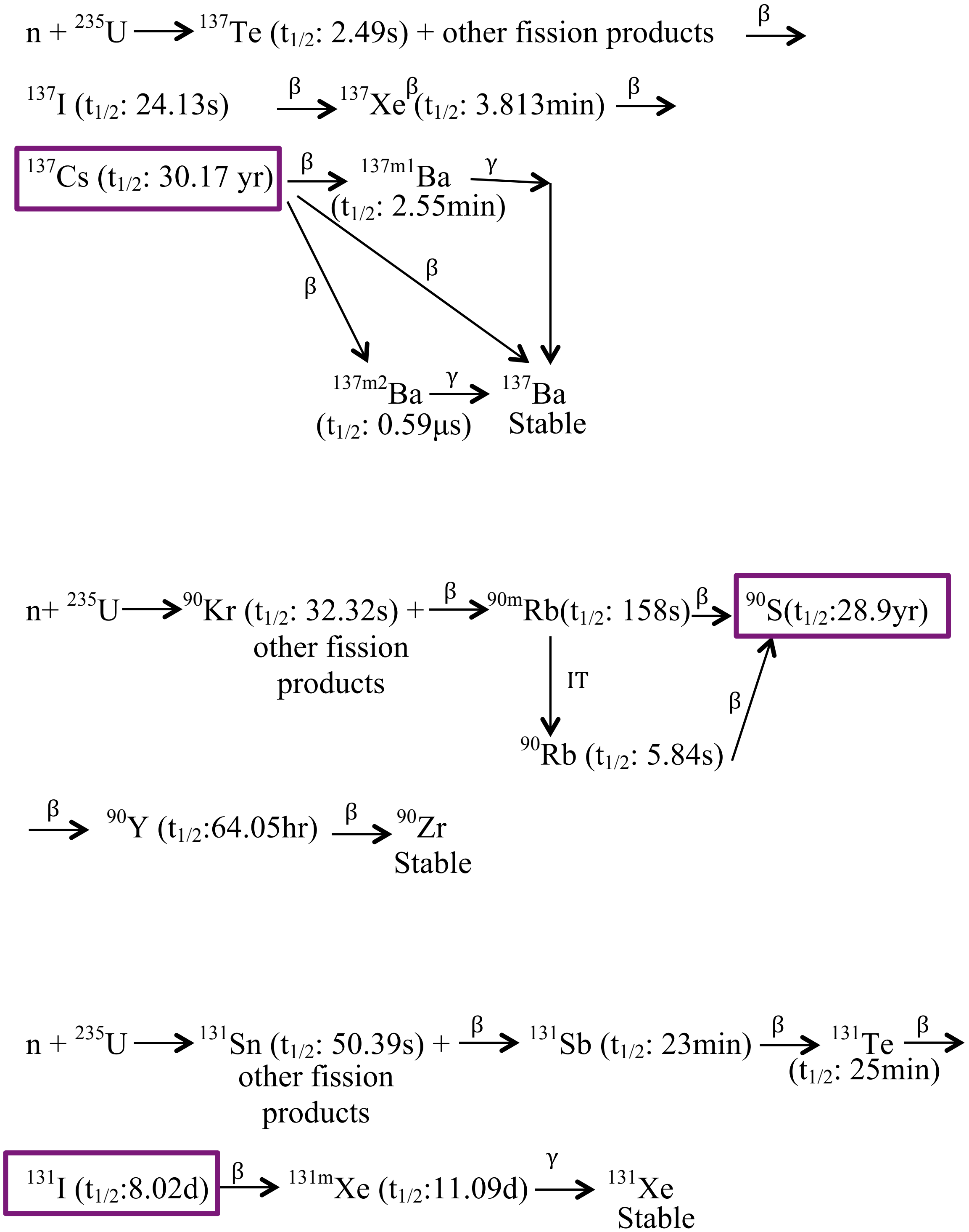

Figure.1. Fission and decay pathway of ^{235}U to produce ^{90}Sr, ^{131}I and ^{137}Cs

APPENDIX

Table App.1- Units for measuring Radioactivity

Quantity	Unit	What is measured	Amount
Activity	Curie (Ci)/Becquerel(Bq)	The number of disintegrations per unit time, 1 Bq= 1 disintegration per second (dps)	1 Ci = 3.7×10^{10}Bq
Absorbed Dose	Rad/Gray (Gy)	The amount of energy absorbed in a mass of matter from radiation, 1Gray = 1J/kg	1Gy = 100 rad
Dose Equivalent	Rem/Sievert (Sv)	The absorbed dose modified by the ability of the radiation to cause biological damage, 1Sv = 1Gy x Quality Factor 1 rem = 1 rad x Quality Factor	1 Sv = 100 rem

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