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Fate of Radionuclides in Wastewater Treatment Plants

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in Civil Engineering

by

Farzaneh Shabani Samgh Abadi

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ABSTRACT OF THE DISSERTATION

Fate of Radionuclides in Wastewater Treatment Plants

by

Farzaneh Shabani Samgh Abadi
Doctor of Philosophy in Civil Engineering
University of California, Los Angeles, 2020
Professor Michael K. Stenstrom, Chair

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 indeed reclamation for potable use is also being practiced. One of the concerns for wastewater reclamation are the contaminants that are not removed by either the wastewater or water treatment processes and this is especially for the case of case of potable reclamation. Radionuclides are rarely a concern in wastewater treatment and reclamation systems, but the recent accident at Fukushima has focused attention on the spread of fission and decay byproduct across farmlands and into drinking water systems.

In addition, recent wildfires in the abundant territory around Chernobyl caused release of long-lived fission products to the atmosphere that had previously been sequestered in the terrestrial system. At this time there is only anecdotal data available on the impact of the fires on the wastewater radioactivity but it is a continuing issue.

An important concern is the fate of radionuclides during wastewater reclamation. In former times, it was assumed that reclamation activities will stop if there is a contamination problem, but with increasing reliance on reclamation, stopping it may have important and perhaps severe effects, including the loss of key industries that use reclaimed water such as petroleum refining. More importantly major cities, including Los Angeles, have aggressive programs for recycling 100% of its wastewater to become a main part of its water portfolio. Therefore, in the future any unexpected interruption in the reclamation programs will have major impacts.

In this dissertation previous observations of the radionuclides in wastewater treatment plants are reviewed and summarized. As part of this research, a variety of wastewater sludge samples were analyzed for radionuclides and results are presented and fate and transport of two radionuclides, ^{131}I and ^{40}K are discussed in detail.

The dissertation of Farzaneh Shabani Samgh Abadi is approved.

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- Sep. 2013-Present, University of California-Los Angeles, Los Angeles, California, USA PhD. in Environmental Engineering: GPA: 3.9 Thesis: Fate of Radionuclides in Wastewater Treatment Plants (Advisor: Dr. M.K. Stenstrom)
- Sep. 2011-Sep. 2013, University of California-Los Angeles, Los Angeles, California, USA M.Sc. in Environmental Engineering: GPA: 3.9 Thesis: Fate of Radionuclides in Wastewater Treatment Plant (Advisor: Dr. M.K. Stenstrom)
- Sep. 2006- Feb. 2011, Sharif University Of Technology, Tehran, Iran B.Sc. in Chemical Engineering: GPA: 3.01 Thesis: Liquid-Liquid Extraction of Zinc

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- Soil Aquifer Treatment to Meet Reclaimed Water Requirements, Water Environment Research, 92(2), September 2019
- Pilot-scale Comparison of Microfiltration/Reverse Osmosis and Ozone/Biological Activated Carbon with UV/Hydrogen Peroxide or UV/Free Chlorine AOP Treatment for Controlling Disinfection Byproducts During Wastewater Reuse, Water Research, 152, April 2019
- Formation of N-nitrosamines during the analysis of municipal secondary biological nutrient removal process effluents by US EPA method 521, Chemosphere, 221, April 2019
- Comparing Industrial and Domestic Discharges as Sources of N-Nitrosamines and Their Chloramine or Ozone-Reactive Precursors, Environmental Science: Water Research & Technology, 5(4), February 2019
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    - Introduction to Environmental Engineering- TA
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    - Food: Lens for Environment and Sustainability- TA
    - Salton Sea, a dying lake- Seminar class- Lecturer

Work Experience- International

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1. Introduction

In southern California water reclamation to increase local water supply is being heavily relied upon. The City of Los Angeles is planning for one hundred percent water recycling by 2035 and reclaimed water is becoming a major portion of the City's water portfolio, both for non-potable and potable reuse. Similar water reclamation practices are either already in place or in planning phases in other states in the US and countries around the world.

The technical and scientific community is becoming more selective in the use of terminology when it comes to "waste"water. Not only is water recognized as an irreplaceable and valuable resource, but also the value of solid "waste" is acknowledged and a great deal of research and effort is being focused on energy and nutrient recovery from biosolids.

Contaminants impose challenges on the reuse practices, mainly in water reclamation. Many contaminants have defined notification limit (NL) and maximum contaminant level (MCL)s that must be met by reclamation practices and contaminants of emerging concern (CEC) are being studied and limits are being developed. Pilot studies on potable and non-potable water reuse continue to evaluate the effectiveness of advanced water treatment (AWT) practices to effectively remove contaminants from reclaimed water. At the same time, the importance of safe discharge of brines and side streams, that are concentrated in contaminants, is recognized and brine management studies are ongoing. Source control is also gaining more attention as part of planning efforts.

One of the less common contaminants in water reclamation and biosolids research are radionuclides. Radionuclides were more commonly observed and studied in wastewater treatment plants during years of atmospheric bomb testing and after Chernobyl. More recently, after Fukushima accident, the environmental fate of radionuclides once again gained attention.

During normal operation of facilities which use radionuclides, from nuclear power plants to medical facilities and laboratories, the release of these contaminates to the environment is well monitored and assumed to be minimal. However, nuclear accidents are inevitable and unpredictable and if they happen, can create major risks for reclamation and reuse practices. Therefore, it is important to understand the fate radionuclides in wastewater systems and whether they are more soluble or have affinity for biosolid.

In the past 60 years, there has been many observations of radioinuclides in the wastewater from previous accidents, bomb fallout and releases from hospitals and industries. [1,3-10,12,14,15-17] This dissertation is a review of the past literature on observations of radionuclides in wastewater treatment plants and research efforts on measuring current levels of radionuclides in wastewater samples at City of Los Angeles Hyperion Water reclamation plant and other wastewater samples. The results are used to develop a mass balance and fate analysis for ^{131}I and ^{40}K, which were measured in all samples.

Chapter 1 includes background information on radiation, basics of radionuclides, sources in the environment and how they appear in wastewater treatment plants. In Chapter 2 previous studies are reviewed to develop a baseline for radioactivity in the wastewater system and an understanding of the impact of spikes in the releases. Chapter 3 describes the testing and methodology, followed

buy results from several observations. Chapter 4 explains the mass balance and fate of ^{131}I and ^{40}K in wastewater samples. Chapter 5 includes discussion of the results and authors main conclusions and suggestions for future research.

2. Background

No matter what the source or concentration of radionuclides is, ionizing radiation is emitted to some extent. The health hazards of ionizing radiation vary greatly among different radionuclides, depending on the type and energy of the emissions as well as the biochemical behavior of the nuclide. Alpha, beta and gamma radiation are the most important ones, with alpha particles known to be the least penetrating and gamma known to be the most penetrating. Table 1 shows the characteristics of each type.

The amount of radiation received from naturally occurring radionuclides or background radiation depends on geographical location and are always present but at different concentrations. Usually they are constant over time and the public need not worry about episodic releases, except in rare cases, such as catastrophic release of mining wastes [18]. Therefore the impact of chronic exposure to naturally occurring radionuclides will not be discussed further in this dissertation.

Radionuclides have several uses in modern society, including medical uses, industrial uses, energy production, and weapons production. As a result, anthropogenic sources to the environment include residues from weapons testing, discharges from medical or scientific uses, industrial use, and power plants. Most radionuclides from these sources are different than natural occurring radionuclides. They include radionuclides produced during the activation process and also fission products. The addition of radionuclides to the environment due to each of these uses must be controlled so that they do not cause health hazards. However, there are some unexpected sources of radionuclides in the environment, such as nuclear power plant accidents. Although several nuclear power plant accidents have occurred which have resulted in improved reactor design and

operation, regulatory agencies are still requiring power plant and reactor designers to anticipate the possibility of accidents and possible prevention methods. Unfortunately, these accidents have happened even in the most technologically developed countries and people are still suffering from the consequences.

The world's increasing population and its increasing use of energy, insures that additional energy supplies must be developed. Fossil fuels, which are still the most common sources for power production, are being used at such high and growing rates that their replacement seems impossible, at least in the near future. Renewable sources such as solar and wind energy are being developed and subsidized to accelerate implementation, but they are unlikely to quickly replace large power plants. Alternative sources such as nuclear energy are still being pursued, and many nations are actively planning to develop nuclear power (including Italy, African countries, North Korea, China, etc.). The environmental safety of using this type of energy needs to be improved and there remains a pressing need to understand and reduce risks associated with nuclear power.

One of the possible problems following the introduction of radionuclides to the environment, which has not been extensively studied, is contamination of wastewaters. This contamination can be caused by radionuclides discharged directly from medical facilities, by atmospheric fallout from accidents or washout of contaminated soils. In former times, contaminating wastewaters was not viewed as a great concern because the wastewaters were segregated from humans for traditional health reasons, and often treatment plants discharged to very large receiving waters were dilution was likely. The advent and increasing dependence on wastewater reclamation is changing this assumption. Figure 2.1 summarized sources and routes to sewer system.

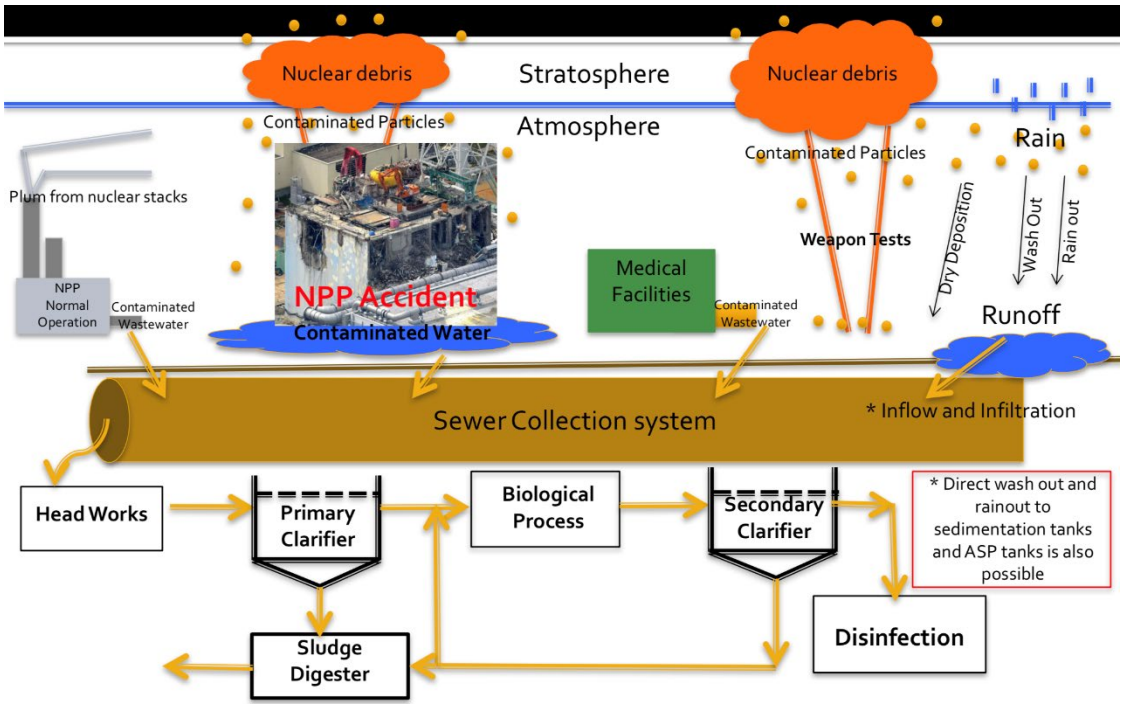


Figure 2.1. Different routes for radionuclides in sewer system

2.1. BASICS OF RADIATION, NUCLEAR POWER PLANTS AND NUCLEAR ACCIDENTS

The decay of unstable nuclides into more stable nuclides is the source of ionizing radiation. The type and energetics of these emissions depends on the characteristics of nucleus. Each radionuclide has its very own set of radiation emissions, and may decay by emissions of alpha, or beta particles with subsequent emission of gamma rays, or a combination of emissions. One type of decay emits alpha particle, which consists of two protons and two neutrons. Alpha particles are the heaviest, most energetic and least penetrating of the various types of ionizing radiation. Radionuclides with large mass numbers (mass number greater than 150, such as ^{222}Rn) are most likely to be alpha

emitters. The energy of alpha particles ranges from 1 MeV to 7 MeV and even the most energetic ones travel only few centimeters in air and can be blocked by a sheet of paper or paint.

The other type of decay emits beta particles, which are basically an energetic e^- (negatron + anti-neutrino) or e^+ (positron + neutrino). Beta particles are generally more penetrating than alpha particles, although beta particles generally cannot penetrate more than few millimeters in human tissues.

Both alpha and beta decay are often accompanied by gamma ray emission. Generally speaking, when the unstable nucleus of a radionuclide decays by emission of alpha or beta particles, the decay leaves the nucleus in an excited state, and the isotope in this state is often called metastable. This excited nucleus will decay to an unexcited state by emission of a gamma ray, which is the most penetrating type of radiation. Gamma ray emission is always observed in coincidence with alpha emission, but may not occur with all beta emission, such as the decay of ^{14}C and ^{3}H. Some of the characteristics of different emissions are listed in Table 2.1.

Table 2.1. Different emissions characteristics

Emission Type	**Characteristics**
Alpha particle ($^4_2$ He)	High energy, Mono-energetic, accompanied by gamma, Least penetrating
Beta$^-$ Particle	Medium to high energy, shares energy with anti-neutrino with $E_{avg} = 1/3(E_{Max})$, more penetrating than alpha with few millimeter range in human tissue, Often but not always accompanied by gamma
Beta$^+$ Particle	Similar to Beta$^-$, a minimum of 1.022 MeV required for this emission and always annihilates with an electron to give 2 x 0.511MeV gamma
Gamma ray	Highly penetrating, high energy, when accompanied by beta likely to interact with matter and produce

Many radionuclides are produced during the normal operation of nuclear reactors in nuclear power plants. These include fission products and activation products. The amount and type of nuclides produced in a nuclear reactor depends on the type of fuel used in the reactor, size of the reactor and the amount of power it produces. However, the results of a nuclear reactor accident depends largely on the amount and types of radionuclides being produced during the accident and the age of the core, as opposed to the normal performance of reactor. The study of such accidents is complicated, in part because of limited information concerning the accidents. Making predictions about types and amounts of nuclides released by accidents is beyond the scope of this dissertation; however, studying previous accidents has provided valuable data for this analysis.

The Chernobyl nuclear power plant and Fukushima Daiichi nuclear power plant accidents are two most disastrous accidents in the history of nuclear energy. The Chernobyl accident is the most disastrous in terms of the total radionuclides release to the environment (11EBq); however, the total amount of radio cesium released by Fukushima accident is comparable to the release from the Chernobyl accident [2,19]. The releases from these two accidents were different because of the difference in the nature of the accidents. Releases from Chernobyl were due to two different processes. The initial release was a large explosive release of radionuclides, including fuel fragments. Alpha emitters are included in fuel fragments. The second release was due to increased temperature within the remaining core. This increase in the temperature was caused by the heat from the decay of fission products and the graphite fires. The increased temperature caused the volatilization and release of fission products (primarily iodine, cesium and strontium).

The Fukushima accident on the other hand, did not suffer a core explosion. The accident mostly resulted in the melting of the fuel. The release of volatile radionuclides was followed by the fuel meltdown. Therefore few fuel fragments and alpha emitters were released by this accident.

Depending on factors such as how well the power plant design meets the “Beyond Design Basis” and the nature of accident itself, the extend of the damage and subsequent hazards following a nuclear accident may be severe to minimal. Several types of fission products can be released following an accident. These radionuclides have half-lives ranging from few milliseconds to a few years. One may define the importance of each of these radioisotopes in terms of environmental hazard based on their half-life, type and energy of the radiation, physical and chemical characteristics of the isotope. Table 2.2 shows the estimated values of the amount of each of radionuclides released to the environment following Chernobyl accident. It is important to note that these are a combination of fission and activation products having a wide range of half-lives. Most of these are beta emitters accompanied by gamma emission; however, there are some larger nuclides from fuel residues, decaying by alpha emission accompanied by gamma emission.

To further discuss the likelihood of observing radionuclides in treatment plants, it is important to know the fate of radionuclides in the environment following such accidents. After their release, radionuclides may appear in the atmosphere or in aquatic and terrestrial ecosystems. Several models have been developed to predict the atmospheric dispersion of radionuclides mostly using the data from nuclear accidents. The most common parameters used in atmospheric dispersion models include advective transport, dispersion, emission, wet and dry deposition and radioactive decay of radionuclides. The term “environmental fate” is used to describe the disposition of chemicals in the environment and their partitions among different compartments in the environment. The environmental fate of radionuclides, like other chemical pollutants, describes how these chemicals are distributed in the environment and final concentrations of them in each environmental compartment. Using environmental transport models, a pathway can be developed

to show how radionuclides make their way to wastewater treatment plants. The study of these processes is beyond the scope of this dissertation; however, it is important to note that the physical and chemical characteristics of radionuclides may cause them to remain close to the accident site, or to be transported to remote locations, spreading well beyond the accident site. Fuel particles and non-volatile nuclides remain close to the accident site while the volatile radionuclides; particularly ^{131}I and ^{137}Cs can spread worldwide.

Table 2.2. Types and amount of Radionuclides Released During the First Days After the Chernobyl Accident, [21] (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	3.06×10^{-6}	~0	~0	
^{134}Cs	2.07 y	0.019	0.43	0.017	5.14	8.2×10^{-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	1.24×10^{-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	3×10^{-5}	6.84×10^{-4}	2.99×10^{-5}	0.009	2.37×10^{-5}	0.09
^{239}Pu	24100 y	2.6×10^{-5}	5.92×10^{-4}	2.6×10^{-5}	0.008	2.6×10^{-5}	0.09
^{240}Pu	6570 y	3.7×10^{-5}	8.43×10^{-4}	3.7×10^{-5}	0.011	3.7×10^{-5}	0.13

The amounts released in accidents and transport properties make $^{89,90}Sr$, ^{131}I and ^{137}Cs, the most important contaminants of concern for wastewater reclamation. The importance of these three

nuclides over other fission and activation products can be explained by their physical, chemical and decay characteristics. Table 2.3 shows some of these characteristics, which include their comparably longer half-lives to other radionuclides, energetics and types of decay and their most likely fate and risks to humans after contact or consumption.

Table 2.3. Summary of characteristics of radionuclides of interest

Nuclide	**Valance**	**Decay mode**	**Half-life**	**Target Organ**	**Activity of 1 gr**
^{89}Sr	+2	β^- (+γ)	50.61 days	Bones	1.1 PBq
^{90}Sr	+2	β^- (No γ)	28.8 years	Bones	5.3 TBq
^{131}I	-1	β^- (+γ)	8.023 days	Thyroid	4.7 PBq
^{134}Cs	+1	β^- (+γ)	2.065 years	Whole Body	49 TBq
^{137}Cs	+1	β^- (+γ)	30.07 years	Whole Body	3.3 TBq

It is notable that nuclear accidents are not the only anthropogenic source or radionuclides in the environment. As an example, ^{131}I is frequently used in medical facilities and these facilities can be one of the main sources of this nuclide in the treatment plants. [1,4,7,15,]

Several radionuclides from different sources with different concentrations have been observed in treatment plants. 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 have medical usage and have been observed in several treatment plants close to medical facilities. ^{99m}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.

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. Radionuclides like ^{137}Cs have negligible decay during the treatment process and will leave the treatment plant as attached to suspended solids or soluble in the effluent.

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 chapter, the observations and results from previous literature on radionuclides in wastewater samples is summarized and discussed in more detail.

3. Review of Previous Literature on Radionuclides in Wastewater Treatment Plants

As part of this research, most of previous studies on radionuclides in wastewater treatment plants were reviewed and results are summarized. This is important in particular to first develop a baseline for which radionuclides have been historically observed at treatment plants and also the impact of accidental releases on doses in wastewater samples. Therefore, results are presented in two separate tables. Table 3.1 presents observations during plants normal operation. Special notes about each study are also included in the table.

It is apparent that some of the more commonly used radionuclides, like ^{131}I are observed in most of the samples. It also appears that ^{131}I concentrations are higher when the sample location was closer to a medical facility, serving a small population, with relatively higher number of patients who need radionuclide-containing procedures. A correlation between peaks in ^{131}I activity in wastewater samples following use of this radionuclide in a close by medical facility has been observed [4,15].^{131}I is also a fission product and if the treatment plant was close to a nuclear power plant, higher activities were observed in wastewater samples. Since ^{131}I has a short half-life of 8.06 days, peaks decay to background levels in 80 days (10 half-lives) or less. ^{7}Be, which is a cosmogenic radionuclide, has also been observed in many of the samples. When precipitation data are available, a correlation between ^{7}Be activity in wastewater samples and precipitation was found.

Table 3.1. Compilation of global observations of radionuclides in wastewater treatment samples between 1960 – 2012. This table excludes spikes due to irregular nuclear events like weapon testing and nuclear accidents

Folsom T.R., Mohanrao, G.J., 1960			
Nuclide Half-life	**Plant Location Biological Process**	**Sample Point (Comment)**	**Activity (Comment)**
^{137}Cs 30.2 years	Hyperion, Playa Del-Rey, CA Partial Secondary	Dried Fertilizer (Dried Digested Sludge)	51.8 Bq/kg (Average of 7 samples over Jan-Feb 1960)
		Raw Sewage	11.8 Bq/kg (Average of 5 samples, each 5 days composites, over Jan-Feb 1960)
Folsom, T. R., et al, 1963 **Note: October 1958 – September 1961: Weapons testing Moratorium. Higher activity of ^{137}Cs at Playa Del-Rey, CA and Portland, OR was linked to food sources.**			
Nuclide Half-life	**Plant Location Biological Process**	**Sample Point**	**Activity**
^{137}Cs 30.2 years	Hyperion, Playa Del-Rey, CA Partial Secondary	Digested Sludge (Discharged to Santa Monica Bay through 7 miles outfall- No dewatering)	58.8 Bq/kg (Average of 103 samples over Mar-Jul 1960)
	Los Angeles County, CA (Carson Plant)- Primary treatment only	Digested Sludge	<2.6 Bq/kg (Average over Nov 1960-Aug 1961)
	Los Angeles County, CA (Carson Plant)- Primary treatment only	Raw Sludge	1.85 Bq/kg (Average over Nov 1960-Aug 1961)
	Atlanta, GA (RM Clayton)-Primary treatment only	Digested Sludge	6.7 Bq/kg (Average over Nov 1960-Aug 1961)
	Atlanta, GA (RM Clayton)-Primary treatment only	Raw Sludge	5.3 Bq/kg (Average over Nov 1960-Aug 1961)
	Baltimore, MD (Back River Plant)- Secondary treatment	Digested Sludge	4.4 Bq/kg (Average over Nov 1960-Aug 1961)
	Baltimore, MD (Back River Plant)- Secondary treatment	Raw Sludge	10.7 Bq/kg (Average over Nov 1960-Aug 1961)
	Iowa City, Iowa (Downtown Plant)-Primary treatment only	Digested Sludge	7.4 Bq/kg (Average over Nov 1960-Aug 1961)
	Iowa City, Iowa (Downtown Plant)-Primary treatment only	Raw Sludge	5.2 Bq/kg (Average over Nov 1960-Aug 1961)
	Portland, OR (Columbia Boulevard)- Primary treatment only	Digested Sludge	111.74 Bq/kg (Average over Nov 1960-Aug 1961)
	Portland, OR	Raw Sludge	58.1 Bq/kg

	(Columbia Boulevard)-Primary treatment only		(Average over Nov 1960-Aug 1961)
	Rochester, MN (Secondary treatment	Digested Sludge	4.8 Bq/kg (Average over Nov 1960-Aug 1961)
	Rochester, MN Secondary treatment	Raw Sludge	3.33 Bq/kg (Average over Nov 1960-Aug 1961)
	San Diego, CA (Point Loma)-Primary treatment only	Digested Sludge	4.81 Bq/kg (Average over Nov 1960-Aug 1961
	San Diego, CA (Point Loma)-Primary treatment only	Raw Sludge	2.59 Bq/kg (Average over Nov 1960-Aug 1961
	San Francisco, CA (South Eastern Plant)-Primary treatment only	Digested Sludge	38.1 Bq/kg (Average over Nov 1960-Aug 1961
	San Francisco, CA (South Eastern Plant)-Primary treatment only	Raw Sludge	28.5 Bq/kg (Average over Nov 1960-Aug 1961
	EMBUD, San Francisco, CA Primary treatment only	Digested Sludge	18.5 Bq/kg (Average over Nov 1960-Aug 1961
	EMBUD, San Francisco, CA Primary treatment only	Raw Sludge	8.5 Bq/kg (Average over Nov 1960-Aug 1961
^{40}K 1.28×10^{10} years	Hyperion, Playa Del-Rey, CA Partial Secondary	Raw Sludge	3.3×10^{-3} g/g (Average of 10 weekly composited samples over Nov 1960-Aug 1961)
	Hyperion, Playa Del-Rey, CA Partial Secondary	Digested Sludge	6.3×10^{-3} g/g (Average of 10 weekly composited samples over Nov 1960-Aug 1961)
	Nine other plants across the USA	Raw Sludge	$2.6 – 9 \times 10^{-3}$ g/g Average: 5.9×10^{-3} g/g (Average of 10 weekly composited samples over Nov 1960-Aug 1961)
	Nine other plants across the USA	Digested Sludge	$2.2 – 7 \times 10^{-3}$ g/g Average: 3.9×10^{-3} g/g (Average of 10 weekly composited samples over Nov 1960-Aug 1961)
^{226}Ra 1.6×10^{3} years	Nine other plants across the USA	Raw Sludge	14.8 – 70.3 Bq/kg Average: 29.6 Bq/kg (Average of 10 weekly composited samples over Nov 1960-Aug 1961)
	Nine other plants across the USA	Digested Sludge	7.4 – 40.7 Bq/kg Average: 0.4 uuc/gr

			(Average of 10 weekly composited samples over Nov 1960-Aug 1961)
^{232}Th 1.4 x 10^{10} years	Hyperion, Playa Del-Rey, CA Partial Secondary	Raw Sludge	<1 – 10.7 x 10^{-6} g/g Average: 4.1 x 10^{-6} g/g (Average of 10 weekly composited samples over Nov 1960-Aug 1961)
	Hyperion, Playa Del-Rey, CA Partial Secondary	Digested Sludge	<1.5 – 16 x 10^{-6} g/g Average: 7.5 x 10^{-6} g/g (Average of 10 weekly composited samples over Nov 1960-Aug 1961)
Erlandsson andMattsson, 1978 **Note: 2 liter samples of digested sludge were counted using a Ge(Li) detector once a week during July 1976- January 1977**			
^{131}I 8.04 days	Malom, Seweden Secondary Treatment	Dewatered Digested Sludge (23% solids)	1.11 – 2.3 Bq/kg
Erlandsson, B., et al, 1982 **Note: dried samples of digested sludge with 3% solids and dewatered digested sludge with 14-15% solids were counted using a Ge(Li) detector during March- September 1981 in a treatment plant 5.8 km (3.6 miles) east of a nuclear power plant**			
^{141}Ce 32.5 days	Borgeby treatment plant, Sweden	Digested Sludge (3% solids)	4.7 – 9.9 Bq/kg
		Dewatered Digested Sludge (14-15 % solids)	4.3 Bq/kg
^{144}Ce 285 days	Borgeby treatment plant, Sweden	Digested Sludge (3% solids)	15 Bq/kg
		Dewatered Digested Sludge (14-15 % solids)	10 Bq/kg
^{103}Ru 39.3 days	Borgeby treatment plant, Sweden	Digested Sludge (3% solids)	5.4 – 18.1 Bq/kg
		Dewatered Digested Sludge (14-15 % solids)	10 Bq/kg
^{106}Ru 373.6 days	Borgeby treatment plant, Sweden	Digested Sludge (3% solids)	7.7 Bq/kg
		Dewatered Digested Sludge (14-15 % solids)	6.8 Bq/kg
^{95}Zr 64 days	Borgeby treatment plant, Sweden	Digested Sludge (3% solids)	13 – 53 Bq/kg
		Dewatered Digested Sludge (14-15 % solids)	9.6 – 28 Bq/kg
^{7}Be 53.2 days	Borgeby treatment plant, Sweden	Digested Sludge (3% solids)	30 – 61 Bq/kg
		Dewatered Digested Sludge (14-15 % solids)	37 – 55 Bq/kg
^{58}Co 70.9 days	Borgeby treatment plant, Sweden	Digested Sludge (3% solids)	<0.6 – 2 Bq/kg
		Dewatered Digested Sludge (14-15 % solids)	Not detected
^{60}Co		Digested Sludge (3% solids)	0.6 – 56 Bq/kg

5.26 years	Borgeby treatment plant, Sweden	Dewatered Digested Sludge (14-15 % solids)	8.5 – 9.1 Bq/kg
^{54}Mn 312.5 days	Borgeby treatment plant, Sweden	Digested Sludge (3% solids)	0.9 – 5.2 Bq/kg
		Dewatered Digested Sludge (14-15 % solids)	1.5 Bq/kg
^{75}Se 119.8 days	Borgeby treatment plant, Sweden	Digested Sludge (3% solids)	4 – 5.6 Bq/kg
		Dewatered Digested Sludge (14-15 % solids)	4 Bq/kg
^{131}I 8.04 days	Borgeby treatment plant, Sweden	Digested Sludge (3% solids)	183 – 1750 Bq/kg
		Dewatered Digested Sludge (14-15 % solids)	91 – 1350 Bq/kg
Erlandsson, B., et al, 1989 Results prior to the Chernobyl accident			
^{7}Be 53.2 days	Lund, Sweden Biotrickling + chemical treatment	Plant Influent	<0.01 – 0.07 Bq/L Average: 0.03 Bq/L 15 samples over July – August 1985
		Plant Effluent	0.03 – 0.04 Bq/L July – August 1985
		Digested dewatered sludge (20-23% solids)	220 – 430 Bq/kg July – August 1985
^{51}Cr	Lund, Sweden Biotrickling + chemical treatment	Plant Influent	0.0227 – 0.136 Bq/L Average: 0.07 Bq/L 15 samples over July – August 1985
		Plant Effluent	0.03 – 0.11 Bq/L July – August 1985
		Digested dewatered sludge (20-23% solids)	200-1100 Bq/kg July – August 1985
^{131}I 8.04 days	Lund, Seweden Biotrickling + chemical treatment	Plant Influent	0.1 – 12.2 Bq/L Average: 1.6 Bq/L 15 samples over July – August 1985
		Plant Effluent	0.29 – 5.31 Bq/L July – August 1985
		Digested dewatered sludge (20-23% solids)	110-1100 Bq/kg July – August 1985
Martin, J.E., 1997 **Note: Samples of 8 hours composite primary sludge, before and after radiotherapy of two patients in a medical facility up stream of the plant**			
^{131}I 8.04 days	Ann Arbor, Michigan Secondary Treatment	Primary Sludge Before radiotherapy	100 Bq/L
		Primary Sludge After radio therapy	192 Bq/L maximum, before leveling off to 100 Bq/L
Fischer, J.W., 2009 **Long term data over 10 years were available for digested sludge and plant effluent. During a short term sample campaign, daily 24 hours composite samples of influent and effluent and daily grab samples of primary sludge were collected and counted. Plant discharges to Weser river, were sediments showed radioisotope contamination. There are**			

3 nuclear power plants on the shore of this river. Isotopes such as ^{40}K and U/Th decay chain members were also detected in the samples; however, activities are not reported in the article.			
^{7}Be 53.2 days	Bermen Seehausen, Germany	Influent	ND
		Effluent	ND
		Primary Sludge	27.6-323 Bq/kg
		Digested Sludge	3.51-556 Bq/kg
^{99m}Tc 6.01 hours	Bermen Seehausen, Germany	Influent	1.18-19 Bq/L
		Effluent	0.067- 3.89 Bq/L
		Primary Sludge	138-6040 Bq/kg
		Digested Sludge	0.19-252 Bq/kg
^{123}I 13.3 hours	Bermen Seehausen, Germany	Influent	ND
		Effluent	ND
		Primary Sludge	2.93-33.6 Bq/kg
		Digested Sludge	ND
^{131}I 8.06 days	Bermen Seehausen, Germany	Influent	0.173-0.856 Bq/L
		Effluent	0.037-0.98 Bq/L
		Primary Sludge	11.8-574 Bq/kg
		Digested Sludge	0.14-201 Bq/kg
^{137}Cs 30.2 years	Bermen Seehausen, Germany	Influent	ND
		Effluent	ND
		Primary Sludge	0.98-3.82 Bq/kg
		Digested Sludge	0.09-32 Bq/kg
^{153}Sm 1.93 days	Bermen Seehausen, Germany	Influent	ND
		Effluent	ND
		Primary Sludge	6.84-165 Bq/kg
		Digested Sludge	ND
Rose, P.S., et al, 2012 **Effluent samples were collected from Stony Brook Water Pollution Control Plant, which serves a thyroid cancer treatment facility. Unfiltered and filtered effluent samples using 0.7 micrometer glass fiber filters were counted. Suspended solids >0.7 micrometer separated from the effluent were also counted.**			
^{131}I 8.06 days	Stony Brook Water Pollution Control Plant- NY Oxidation Ditch	Unfiltered effluent	1.8-217 Bq/L 77 days between June 2006-March 2009
		Filtered effluent	2.5-227 Bq/L 46 days between January 2007-March 2009
		Suspended solids in the effluent >0.7 micrometer	6.1×10^{4}- 2.8×10^{6} Bq/kg 36 days between March 2007-March 2009

Some of the naturally occurring radionuclides were also observed in many of the samples, although their activities were not always reported, since it is assumed that these activities will remain relatively constant. However, it is important to keep a track of their baseline levels in background studies and they can be useful in wastewater tracing studies. An example of such radionuclide is

^{40}K. Authors have recently measured and studied the fate of this radionuclide, which has been previously reported to have similar fate to that of ^{137}Cs.

Table 3.2 summarizes observations following a known spike in environmental release of radionuclides either from a nuclear accident or bomb testing. There is an obvious increase in the activity of some of the background radionuclides, like ^{131}I, which are also fission products following these events and a wide variety of fission products were observed comparing to background levels and depending on the radionuclides half-lives, their activities eventually decayed to background levels. Where the comparison of the before and after spike release is possible, it is important to note that the levels of non-fission products remains relatively unchanged.

The amount and types of radionuclides that can end up in a wastewater treatment plant after spike in releases depends one a variety of factors, including the physical and chemical characteristics of the radionuclide. Nevertheless, elevated levels in wastewater sample are expected. Some of these have short half lives and will decay quickly before exiting the plants and some will decay to background levels in longer periods of times. Whether these spike in releases can cause a safety issue for plant workers and operators depends on many factors, including types of radionuclides and activities.

Table 3.2. Spikes in radioactivity in samples from wastewater treatment plant following irregular nuclear activities

Imhoff K.R., et al, 1988 **Note: Sludge samples from treatment plants in Germany were measured for gamma activities and gross beta between 1960-1986, after the Chernobyl accident, until levels decreased to background. These measurements also captured the impact of nuclear weapon testing**			
^{131}I 8.04 days	Duisburg- Kasslerfeld	Digested Sludge	1600 - <10 Bq/kg between 5/22/1986 to 9/30/1986
	Arnsberg	Digested Sludge	3370 to <10 Bq/kg between 5/22/1986 to 9/25/1986
	Hagen	Digested Sludge	7180 to <10 Bq/kg between 5/22/1986 to 9/9/1986
	Warstein	Digested Sludge	2570 to <10 Bq/kg between 5/27/1986 to 9/5/1986
	Essen- Burgaltendorf	Digested Sludge	2520 to <10 Bq/kg between 5/28/1986 to 9/30/1986
	Meinerzhagen	Digested Sludge	1370 to <10 Bq/kg between 5/6/1986 to 9/10/1986
	Hattingen	Digested Sludge	1034 Bq/kg on 6/10/1986
	Bochum - Olbachtal	Centrifuged primary, WAS and digested sludge on 1:1:1 ration	3400 to <10 Bq/kg between 5/12/1986 to 10/3/1986
	Witten	Digested Sludge	420 Bq/kg on 6/10/1986
	Ruthen	Digested Sludge	3230 to <10 Bq/kg between 6/12/1986 to 9/24/1986
^{137}Cs 30.2 years	Duisburg- Kasslerfeld	Digested Sludge	2700 - 160 Bq/kg between 5/22/1986 to 9/30/1986
	Arnsberg	Digested Sludge	10380 to 900 Bq/kg between 5/22/1986 to 9/25/1986
	Hagen	Digested Sludge	3830 to 1190 Bq/kg between 5/22/1986 to 9/9/1986
	Warstein	Digested Sludge	5620 to 1300 Bq/kg between 5/27/1986 to 9/5/1986
	Essen- Burgaltendorf	Digested Sludge	6990 to 800 Bq/kg between 5/28/1986 to 9/30/1986
	Meinerzhagen	Digested Sludge	6170 to 1710 Bq/kg between 5/6/1986 to 9/10/1986
	Hattingen	Digested Sludge	4850 Bq/kg on 6/10/1986
	Bochum - Olbachtal	Centrifuged primary, WAS and digested sludge on 1:1:1 ration	2600 to 310 Bq/kg between 5/12/1986 to 10/3/1986
	Witten	Digested Sludge	5900 Bq/kg on 6/10/1986
	Ruthen	Digested Sludge	9150 to 3930 Bq/kg between 6/12/1986 to 9/24/1986
Residual beta activity	Treatment plants of the Ruhr River Association	Sludges*	Samples analyzed between 1960-1968 and obvious drop observed in activities >3500 Bq/kg after nuclear weapon tests stopped in 1963. All activities dropped to <900 Bq/kg by 1966. High values of 8800 and 10100 Bq/kg observed in 1962 and 1963

			were similar to those observed after Chernobyl
DurhamJoshi,1979 **Effluent and sludge samples from two treatment plants close to Lake Ontario were counted using Ge(Li). It is not specified in the paper that what kind of sludge was sampled. Radioisotopes are a mixture of naturally occurring, medically used and fission products from weapon testing.**			
^{144}Ce 285 days	Hamilton Plant	Dried Sludge	46.2 Bq/kg
		Effluent	ND
	Dundas Plant	Dried Sludge	25 Bq/kg
		Effluent	ND
^{141}Ce 32.5 days	Hamilton Plant	Dried Sludge	5.4 Bq/kg
		Effluent	ND
	Dundas Plant	Dried Sludge	ND
		Effluent	ND
^{226}Ra 1622 years	Hamilton Plant	Dried Sludge	37.9 Bq/kg
		Effluent	ND
	Dundas Plant	Dried Sludge	23.4 Bq/kg
		Effluent	ND
^{228}Th 1.9 years	Hamilton Plant	Dried Sludge	8.7 Bq/kg
		Effluent	ND
	Dundas Plant	Dried Sludge	10.5 Bq/kg
		Effluent	7.4×10^{-4} Bq/L
^{75}Se 120 days	Hamilton Plant	Dried Sludge	ND
		Effluent	ND
	Dundas Plant	Dried Sludge	2.4 Bq/kg
		Effluent	ND
^{51}Cr 27.8 days	Hamilton Plant	Dried Sludge	100 Bq/kg
		Effluent	0.05 Bq/L
	Dundas Plant	Dried Sludge	ND
		Effluent	ND
^{131}I 8.04 days	Hamilton Plant	Dried Sludge	1.7 Bq/kg
		Effluent	ND
	Dundas Plant	Dried Sludge	6.8 Bq/kg
		Effluent	ND
^{125}Sb 2.7 years	Hamilton Plant	Dried Sludge	5.3 Bq/kg
		Effluent	ND
	Dundas Plant	Dried Sludge	ND
		Effluent	ND
^{7}Be 53.2 days	Hamilton Plant	Dried Sludge	82.4 Bq/kg
		Effluent	8.5×10^{-3} Bq/L
	Dundas Plant	Dried Sludge	30.6 Bq/kg
		Effluent	ND
^{103}Ru 39.3 days	Hamilton Plant	Dried Sludge	8.5 Bq/kg
		Effluent	ND
	Dundas Plant	Dried Sludge	ND
		Effluent	ND
^{106}Ru 1 year	Hamilton Plant	Dried Sludge	35.2 Bq/kg
		Effluent	ND
	Dundas Plant	Dried Sludge	ND
		Effluent	ND
^{137}Cs	Hamilton Plant	Dried Sludge	7.8 Bq/kg

30.2 years		Effluent	9.2×10^{-4} Bq/L
	Dundas Plant	Dried Sludge	3.7 Bq/kg
		Effluent	ND
^{95}Zr 64 days	Hamilton Plant	Dried Sludge	9.4 Bq/kg
		Effluent	ND
	Dundas Plant	Dried Sludge	ND
		Effluent	ND
^{95}Nb 35.1 days	Hamilton Plant	Dried Sludge	13.5 Bq/kg
		Effluent	ND
	Dundas Plant	Dried Sludge	ND
		Effluent	ND
Folsom, T.R., et al, 1963 **Soviet Union resumes weapon testing in September 1961. Fission products were observed in raw sludge samples from Hyperion, CA and in Portland, Oregon**			
Hyperion-CA	September 1961-Before testing started	Raw Sludge	^{65}Zn, ^{137}Cs and ^{40}K
	November 1961-After testing started	Raw Sludge	^{144}Ce, ^{106}Ru, ^{106}Rn, ^{137}Cs, ^{95}Zr, ^{95}Nb, ^{65}Zn, ^{60}Co
Portland- OR	July 1961-Before testing started	Raw Sludge	^{131}I, ^{137}Cs and ^{60}Co
	September 1961-After testing started	Raw Sludge	^{144}Ce, ^{106}Ru, ^{106}Rn, ^{137}Cs, ^{95}Zr, ^{95}Nb, ^{65}Zn, ^{40}K
Erlandsson and Mattsson , 1978 **Note: 40 days after a 200 kton nuclear weapon was tested in China**			
^{131}I 8.04 days	Malom, Seweden Secondary Treatment	Dewatered Digested Sludge (23% solids)	15.5 Bq/kg
Erlandsson, B., et al, 1989 **Results after the Chernobyl accident- Sewers are combined**			
^{7}Be 53.2 days	Lund, Sweden Biotrickling + chemical treatment	Plant Effluent	0.015 – 0.037 Bq/L
		Digested dewatered sludge (20-23% solids)	190 – 300 Bq/kg
^{51}Cr 27.8 days	Lund, Sweden Biotrickling + chemical treatment	Plant Effluent	0.06 – 0.16 Bq/L
		Digested dewatered sludge (20-23% solids)	290 – 650 Bq/kg
^{106}Ru 1 year	Lund, Sweden Biotrickling + chemical treatment	Plant Effluent	<0.01- 0.065 Bq/L
		Digested dewatered sludge (20-23% solids)	5.5 – 32 Bq/kg
^{131}I 8.04 days	Lund, Sweden Biotrickling + chemical treatment	Plant Effluent	0.6 – 20 Bq/L
		Digested dewatered sludge (20-23% solids)	290 – 2400 Bq/kg
^{134}Cs	Lund, Sweden	Plant Effluent	<0.01 – 0.012 Bq/L

2.06 years	Biotrickling + chemical treatment	Digested dewatered sludge (20-23% solids)	17 – 40 Bq/kg
^{137}Cs 30.2 years	Lund, Sweden Biotrickling + chemical treatment	Plant Effluent	<0.01-0.023 Bq/L
		Digested dewatered sludge (20-23% solids)	50 – 160 Bq/kg
Fischer, H. W., Yokoo, Y., 2014 **Sludge samples (does not identify sludge type and characteristic) from Fukushima city wastewater treatment plant (WWTP) was sampled and counted between end April 2011- end December 2013. Activities of ^{137}Cs and ^{131}I are reported. ^{134}Cs was also observed and reported at 1:1 with ^{137}Cs. Sewage system is separated. However, isotopes concentrations correlate with precipitation**			
^{131}I 8.04 days	Fukushima city WWTP-Japan	Sludge	9 – 1000 Bq/ kg
^{137}Cs 30.2 years	Fukushima city WWTP-Japan	Sludge	70 – 6000 Bq/kg

Biosolids can also carry elevated levels of radionuclides. Whether it is possible to store the biosolids for long enough that the radionuclides decay, before they are released to the environment also depends on types and activities and also plant's capabilities to handle and store solids.

In AWT plants, the radionuclides most likely will concentrate in the brine streams and require more care in handling.

The impact of spike in release of radionuclides in wastewater treatment plants in modern time, with heavy reliance on water reclamation programs for potable or non-potable reuse needs to be looked into further.

4. Testing, Methodology and Results

4.1. Instruments and Equipment Setup

This research was initiated by a simple lab set up to measure radioactivity in different samples and modifications were made later on to improve the analytical capabilities. The initial set up included a Ludlum 2200 single channel analyzer, 6 cm in x 6 cm in NaI scintillation detector and lead box with approximately 10 cm of lead thickness on each side as a shield from background noise. The configuration of the lead box was so that the NaI detector had to be removed from the top of the box in order to move the samples inside and outside of the box. Figure 4.1 shows an image of the lead box. Three isotopes were used to calibrate the single channel analyzer at specific energy ranges: ^{137}Ca, ^{22}Na and ^{60}Co each at 1 uCi.

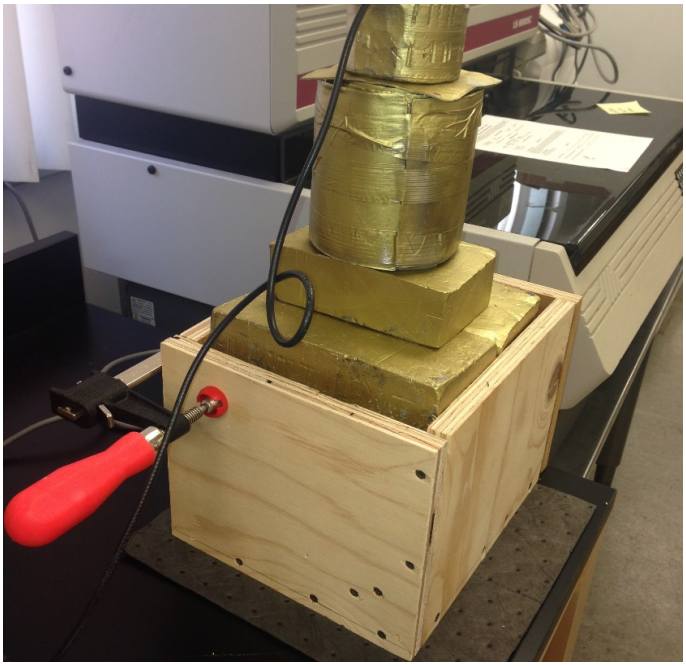


Figure 4.1. First lead box

Thought with this set up it was possible to detect gamma radiation from known sources, with gamma energies in the range the single channel analyzer was calibrated at, it was inadequate for two main reasons for the purpose of this study. First, the main goal of this research was to measure and document observations of all varieties of radionuclides in wastewater samples. These can include radionuclides with short and long half-lives. The time delay caused by the inadequacy of a single channel analyzer to detect multiple gamma energies at the same time could result in the decay of radionuclides like ^{131}I with short half-lives, before they were even counted. Second, due to the lead box configuration, having to move the detector in between each sample analysis would result in inconsistent geometry of detector relative to the samples, which is not ideal in gamma spectroscopy.

In order to overcome these and enhance analytical capabilities, the testing set up was improved by making the following changes:

a) A new 10 cm x 10 cm NaI scintillation detector
b) Changing the Ludlum 2200 single channel analyzer with an Ortec digiBASE multichannel analyzer (MCA), compatible with the 10 cm x 10 cm NaI detector
c) MAESTRO MCA application, version 7.01
d) Enhanced the lead box configuration by first, adding more lead bricks and making a bigger space inside the box to be able to fit larger sample volume for each counting. This modification made it possible to fit more than twice the amount of sample in the lead box compared to the initial set up.

Second, the wall thickness on each side of the box was increased to at least 20 cm compared to the initial 10 cm.

Lastly, the detector was set at a fixed position, facing the sample, with face of the detector 10 cm away from the sample. A side opening was created in order to have disturbing the position of the detector.

Figure 4.2 show an overview of the equipment set up after the improvements were made.

Figure 4.2. Improved set up

4.2. System Calibration and Background Counting

The system was calibrated before analyzing each sample using the three standards and KCl table salt as a source for ^{40}K. In order to create a standard from the table salt, 360 grams of salt was packaged in 10 cm x 10 cm circular plastic containers, which were used to package all the other

samples and the expected activity was calculated by assuming 0.012 percent of total potassium in the salt is ^{40}K, similar to what is expected in nature. Table 5 is a summary of standards, their activity and gamma energies.

Before counting any standard or sample, a background counting was performed for the same duration of time that the sample or the standard was counted. For calibrations, ten minutes background counting and ten minutes counting of the standards were performed.

Once calibration is done, MAESTRO can analyze the gamma spectrum to identify peaks and total counts in each peak. This information was used after counting the standards and compared with the expected activities listed in Table 4.1 to develop an efficiency curve for the detector. Figure 4.3 presents the efficiency curve.

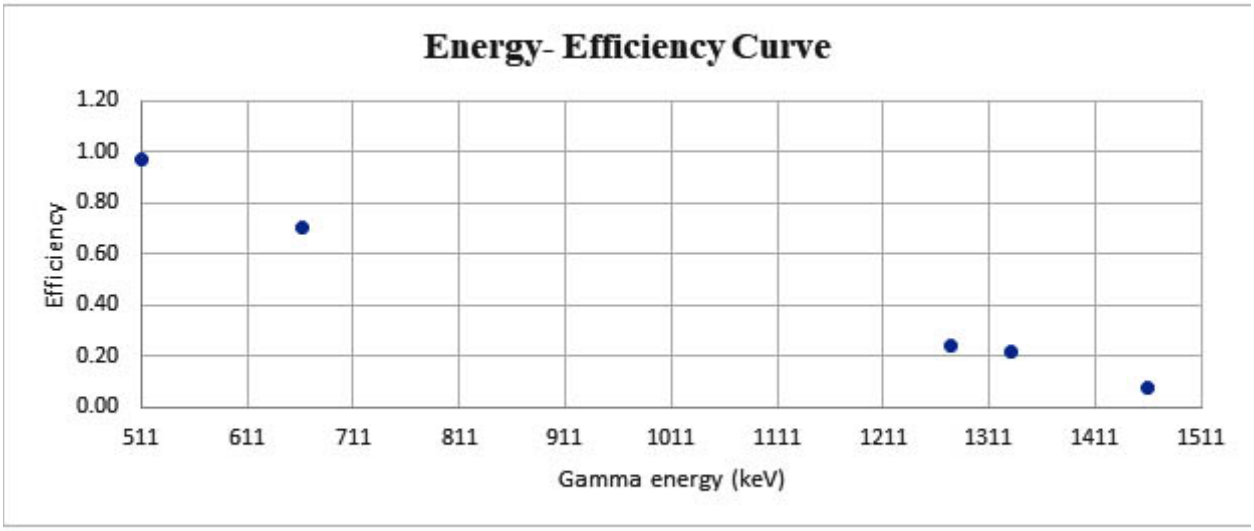


Figure 4.3. Energy efficiency curve

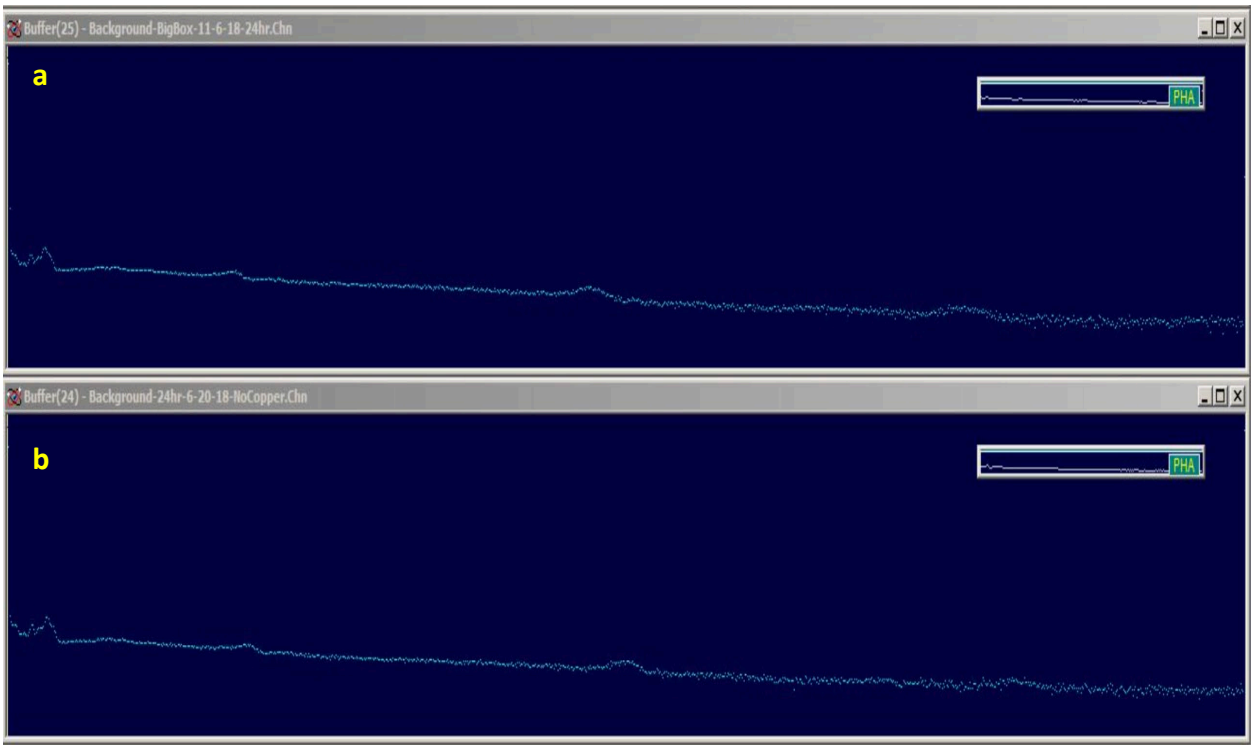
Table 4.1. Standards, activities and gamma energies

Standard	Activity (Bq*)	Gamma energy (keV)
^{40}K	590[1]	1,461
^{60}Co	3,700	1,173
		1,332
^{137}Cs	3,700	663
^{22}Na	3,700	1274
		511

*Bq = disintegration per second (cps)

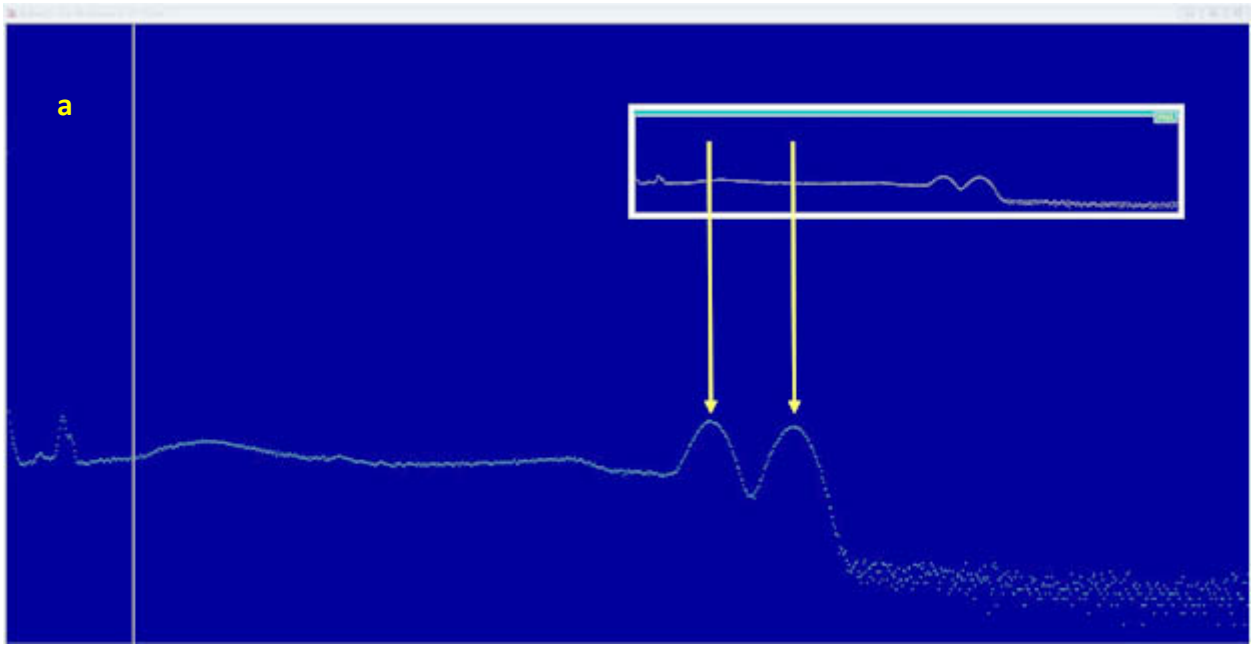
1) % radioactive = 0.012%, 360 grams of KCl salt = 360/75.5 or 4.83 moles, 4.83 x 0.00012= 5.7 x 10^{-4} moles ^{40}K or 3.43x10^{19} atoms, decay coefficient= 0.693/ (1.28 x 10^{9} x 365 x 24 x 3600)= 1.72 x 10^{-17}/s, Activity = 3.43x10^{19} x 1.72 x 10^{-17} = 590 cps

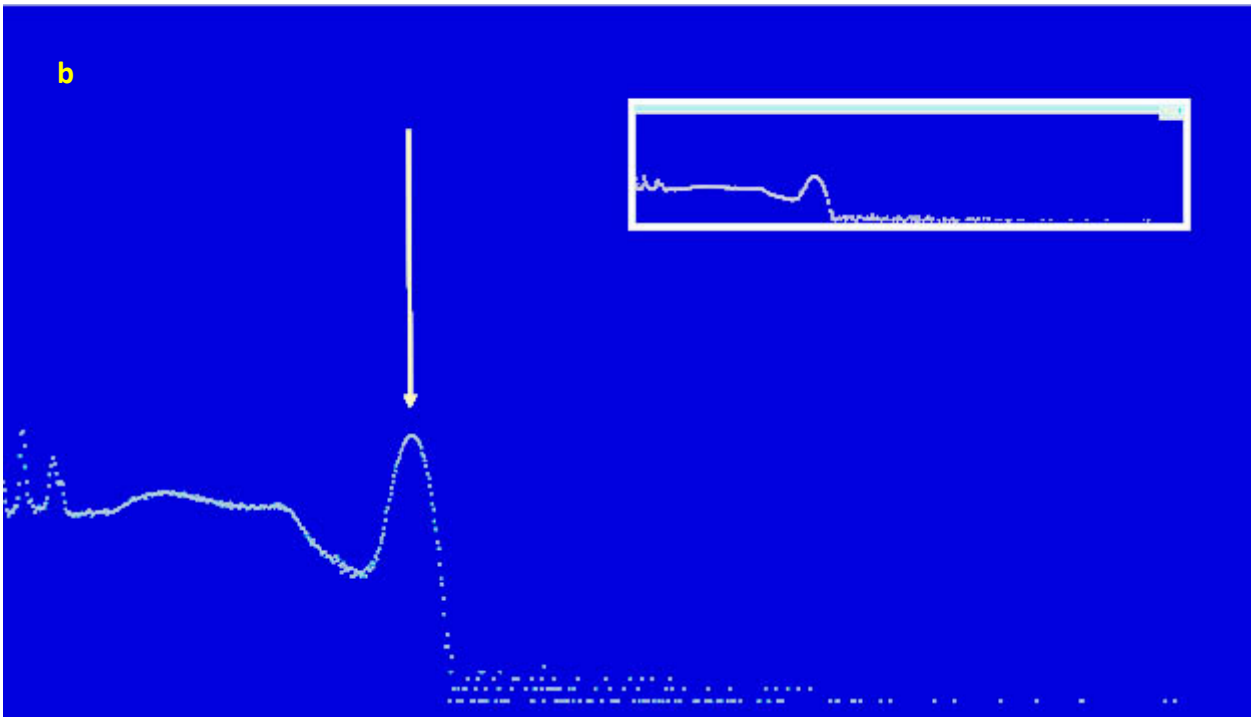
Figures 4.4-a and b present examples of 24 hours background counting for the small and the large lead boxes. The total counts from all 1024 channels are comparable in both cases and on average less than 8 percent different.



Figures 4.4- a and b examples of 24 hours background counting for the small and the large lead boxes

Figures 4.5-a, b, c and d are examples of ten minutes counting of the ^{60}Co, ^{137}Cs, ^{40}K and ^{22}Na standards respectively for calibration.





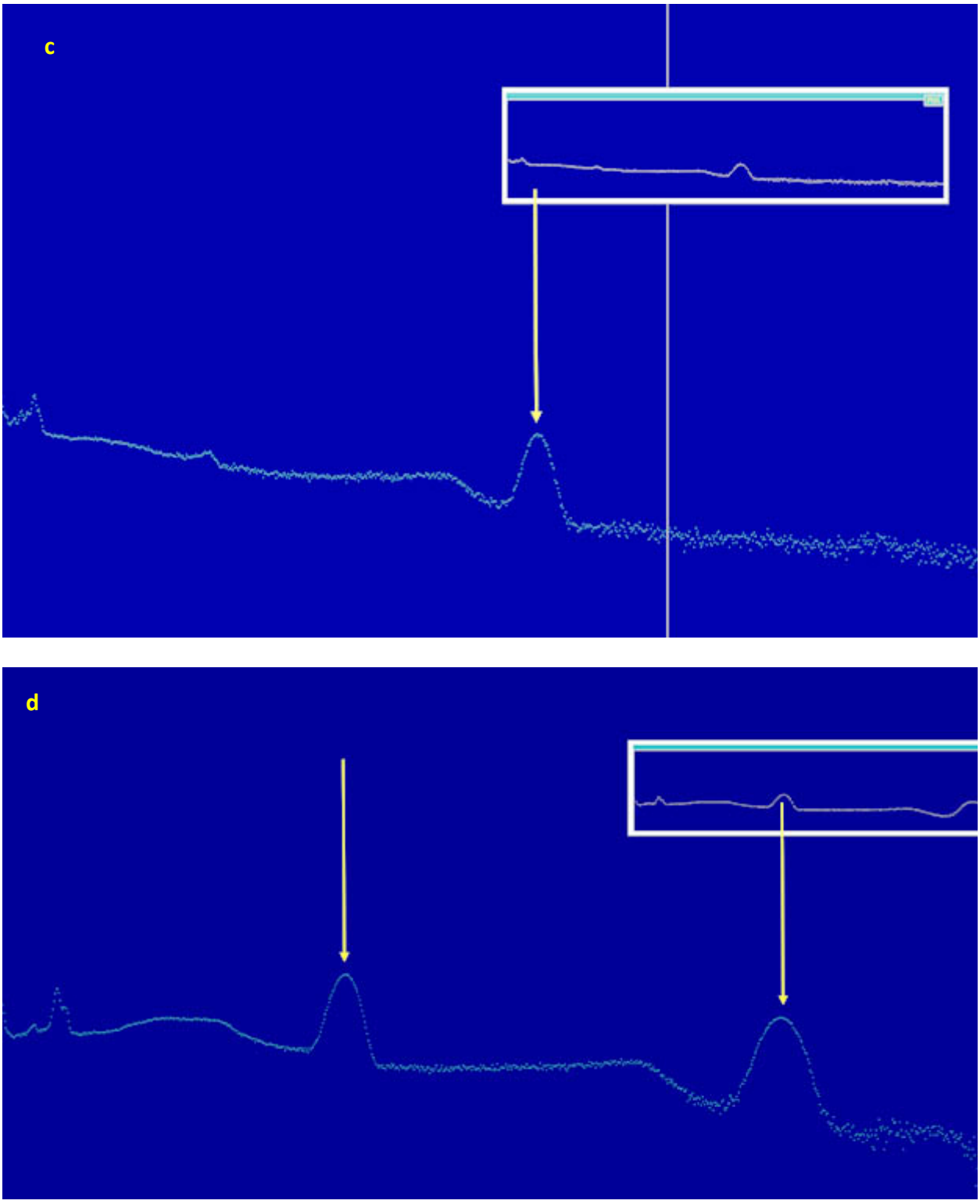


Figure 4.5-a to d: ten minutes counting of the a: ^{60}Co, b: ^{137}Cs, c: ^{40}K and d: ^{22}Na standards

4.3. Sample description and Results Summary

The focus of this research was mainly on the sludge samples from the City of Los Angeles Hyperion Water Reclamation Plant (HWRP), which is the largest of the four water reclamation plants operated by Los Angeles Satiation and the Environment (LASAN). The HWRP treatment process includes headworks, primary settling, high purity oxygen activated sludge (HPOAS) biological treatment and final settling, currently treating 260 MGD average dry weather flow.

HWRP process the biosolids on site. Sludge from primary and secondary settling tanks are separately thicken by centrifuge. A mixture of thickened primary (TPS) and thickened waste activated sludge (TWAS) mixed with primary sludge for dilution enters the digesters. The two sludge streams have 4.2 and 4.75 percent total solids respectively. The detention time in the digestion process is about 16 days and average temperature of 129 degree F. Digested sludge has 2.1 percent total solids and enters the digester screening facility after the digestion process, where it is dewatered to 26-27 percent total solids. Figure 4.6 show a simple process flow diagram for the solid processing at HWRP.

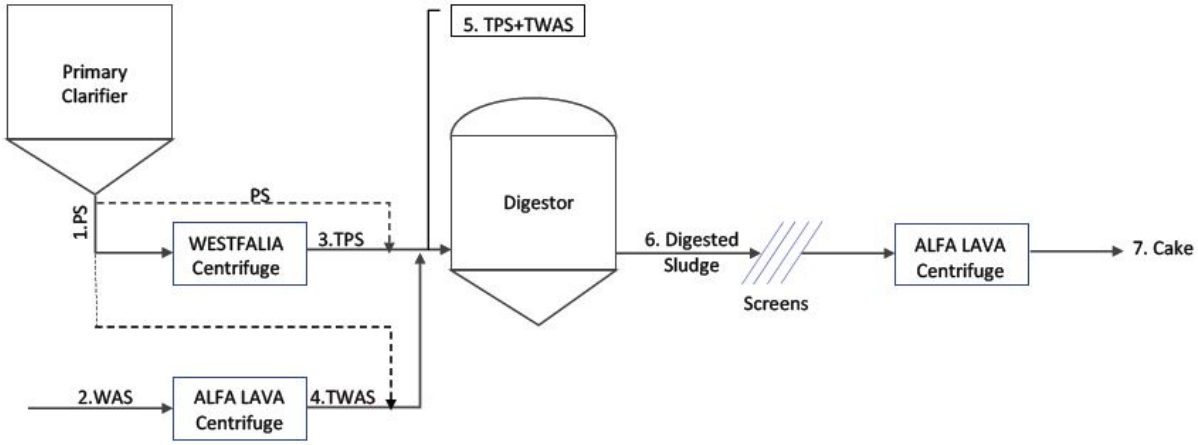
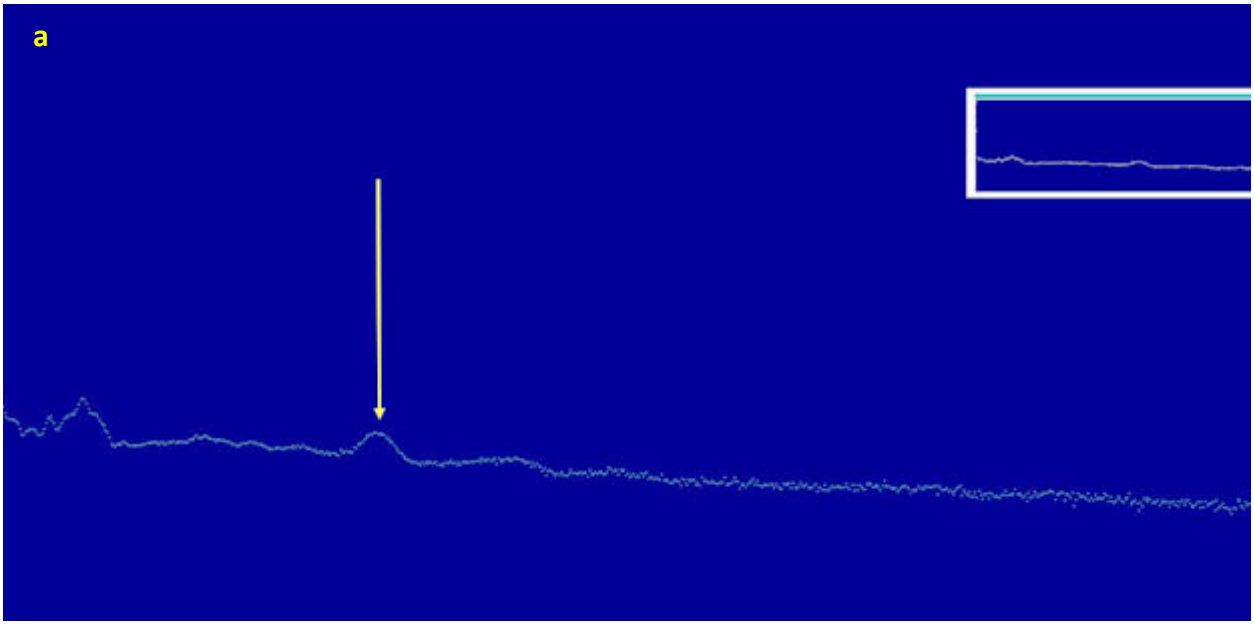


Figure 4.6. Process flow diagram for HWRP solid processing

The dewatered digested sludge (cake) was the first sample that was collected from HWRP. This sample was analyzed immediately at UCLA using the instruments and equipment described in section 4.1. At the same time, a portion of the sample was sent to UCI and analyzed at the nuclear engineering laboratory, using a HPGe detector. Figure 4.7-a and b presents results of the analysis at UCLA and UCI respectively.



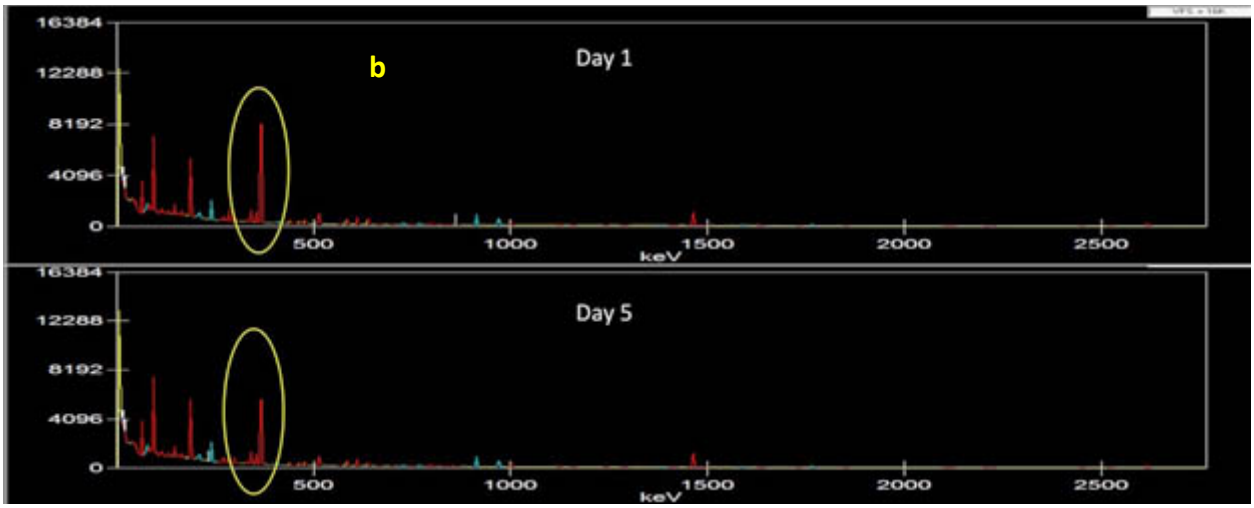


Figure 4.7. Results of the analysis of the first cake sample from HWRP at a: UCLA and b: UCI

Although a peak at nearly 360 keV was apparent, it was not large enough to confirm the type of radionuclide. Analysis of the same sample using a HPGe detector, which has a higher resolution compared to the NaI detectors has a better outcome The sample was counted twice and four days apart. The presence of ^{131}I in this sample was evidenced by the peaks at 365 keV, 284 keV, and 637keV and confirmed based on the decay between the two counting.

The second and third samples collected from HWRP included those from points 3-4 and 6-7, identified on Figure 4.6. It was necessary to increase the activity in the amount of sample that could be fit inside the lead box by concentrating the radionuclides in a smaller volume. Concentration of radionuclides is proportional to their activity in the sample. In order to reduce the sample volume while retaining the radionuclides that are both in the solid and liquid phase, the second and third samples set of samples were dried under sunlight, on the engineering building roof. The second set of samples were spread over shallow pans, dried and counted 3 weeks after receipt at the lab. Dried solids were then ground and packaged inside the sample containers.

Between 300-700 gr of dried solids were packaged and counted multiple times to validate the radionuclides identified in the samples based on their decay rate. Total counts were corrected for ^{131}I decay for the time laps between sample collection at Hyperion Water Reclamation Plant and, for the digested sludge, for sludge detention time in the digestion process. The counting time for ^{131}I was corrected using Hoffman and Van Camerik method. ^{40}K was assumed to have not decayed due to its long half-life.

The third set of samples had the longest time laps between receipt of samples in the lab and counting due to the larger volume of the samples, the wet season in Los Angeles interrupted the natural drying process for 1 month and colder temperatures slowed down the drying process. Overall it took 95 days for these samples to dry completely. Once the samples were completely dry, 390 – 960 gr of dried solids were ground and packaged in the sample containers and counted. At this point the ^{131}I had decayed completely and only ^{40}K was observed.

All samples were counted for 12 to 48 hours multiple times, with 1-2 days and up to 24 months in between, in order to confirm the radionuclides that were identified in the samples based on their decay rates. Figure 4.8 shows an example of the activity of ^{131}I in a thickened primary sludge sample (TPS) analyzed on four different days.

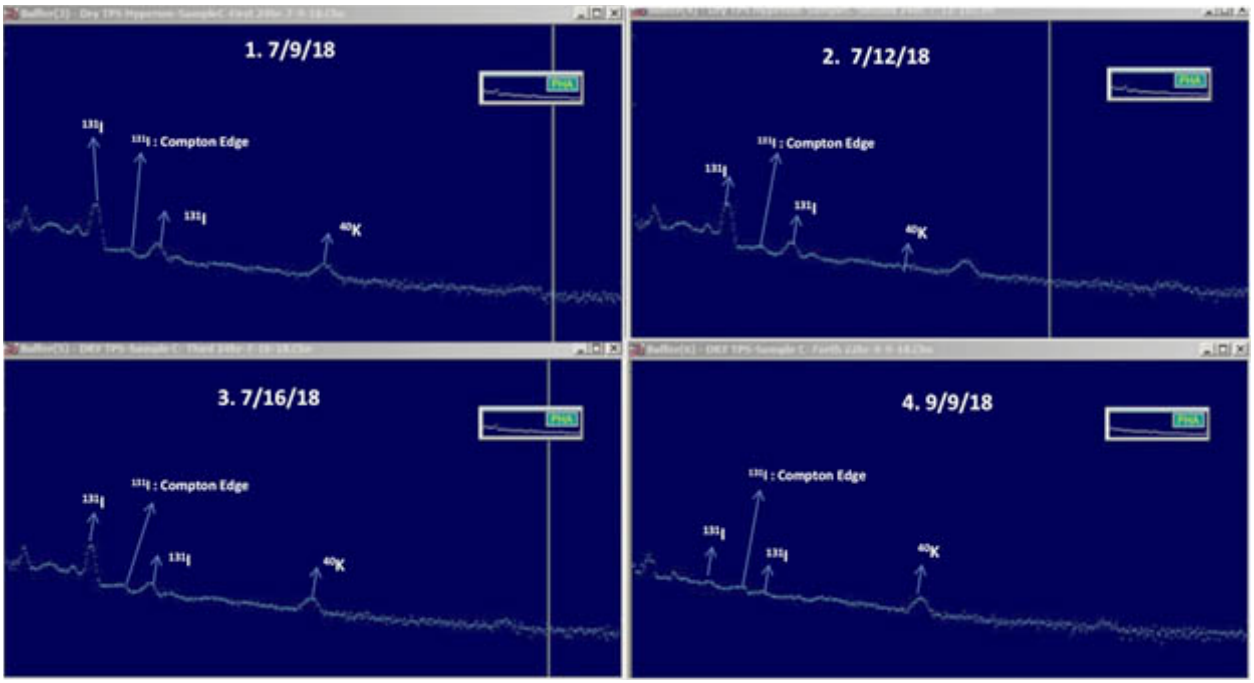


Figure 4.8. Example of analysis of a TPS sample

In order to confirm the radionuclide identified in the sample based on the decay rate, the following estimation method was used:

I-131 half Life: 8.04 days Eq (4.1)

First count on 7/9/18 for 24 hours: 190,000 total counts Eq (4.2)

Second count on 7/12/18 for 24 hours counting: 149,500 total counts Eq (4.3)

Third count on 7/16/18 for 24 hours counting: 104,500 total counts Eq (4.4)

Forth count on 9/9/18 for 24 hours counting: 860 total counts Eq (4.5)

$N_t = N_0 e^{-\lambda t}$ Eq (4.6)

$\lambda = 0.693 / 8.04$ days Eq (4.7)

$N_{7/12/18} = N_{7/9/18} e^{-\lambda t} = 140{,}500$ total Eq (4.8)

$N_{7/16/18} = N_{7/12/18} e^{-\lambda t} = 105{,}902$ total Eq (4.9)

$N_{9/9/18} = N_{7/16/18} e^{-\lambda t} = 830$ total Eq(4.10)

These results confirm ^{131}I in the TPS sample based on its decay rate. Similar analysis was performed for all other samples.

Beside the HWRP sludge samples, an incinerated, combined primary and waste activated sludge sample from a northern California wastewater treatment plant operating at low solids retention time (SRT) was analyzed. Also two different types of biosolids-derived fertilizer/soil amendments were also analyzed. These samples were generally more concentrated because of incineration or more complete dewatering. Results from the sample counting are summarized in Table 4.2.

Table 4.2. Summary of the results

Sample type	**Radionuclide**	**Half life**	**Activity**
Incinerated Sludge	^{141}Ce	32 days	1.3 Bq/Kg incinerated sludge
	^{40}K	1.28×10^{10} years	7.3 g/kg incinerated sludge
	^{51}Cr	27.7 days	0.4 Bq/kg incinerated sludge
Struvite-based Fertilizer	^{40}K	1.28×10^{10} years	5.3 gr/kg fertilizer
Biosolids- Fertilizer	^{40}K	1.28×10^{10} years	6.1 gr/kg fertilizer
Hyperion Thickened Primary Sludge	^{131}I	8.04 days	78.9 Bq/kg
	^{40}K	1.28×10^{10} years	0.22 – 0.28 gr/kg
Hyperion Thickened Waste Activated Sludge	^{131}I	8.04 days	69.3 Bq/kg
	^{40}K	1.28×10^{10} years	0.25 – 0.31 gr/kg
Hyperion digested sludge	^{131}I	8.06 days	70.4 Bq/kg
	^{40}K	1.28×10^{10} years	0.32 – 0.38 gr/kg
Hyperion dewatered digested sludge	^{131}I	8.04 days	34.3 Bq/kg
	^{40}K	1.28×10^{10} years	1.3 – 1.6 gr/kg

These results are used in Chapter 4 to develop a mass balance for ^{131}I and ^{40}K.

5. Mass Balance and Fate of ^{131}I and ^{40}K in Wastewater samples

Table 4.2 summarized the results from analyzing the sludge samples from HRWP, biosolids fertilizers and the incinerated sludge samples. The HWRP results are used to develop a solid mass balance and activity balance in order to learn the fate and segregation of ^{131}I and ^{40}K, which consistently appeared in all samples between the solid and liquid phase. Figure 5.1 presents flow and mass rate throughout the solid processing facility at HWRP based on the plant's monthly performance reports (MPR).

The percentage solid difference between incoming sludge to the digesters and dewatered digested sludge, 4.5 versus 26- 27 percent, was adequate to do a solid versus liquid affinity analysis. These percentages were confirmed using the total mass of the sludge that was air dried and the mass of the remaining dried solids

In order to develop the solid mass balance, it was assumed that the non-volatile solids mass (non VS) is equal to the difference between the total solid mass (TS) and total volatile solids (VS) and remains constant over the digesters. This assumption is validated by comparing the mass per days for each of these solids from the MPR results.

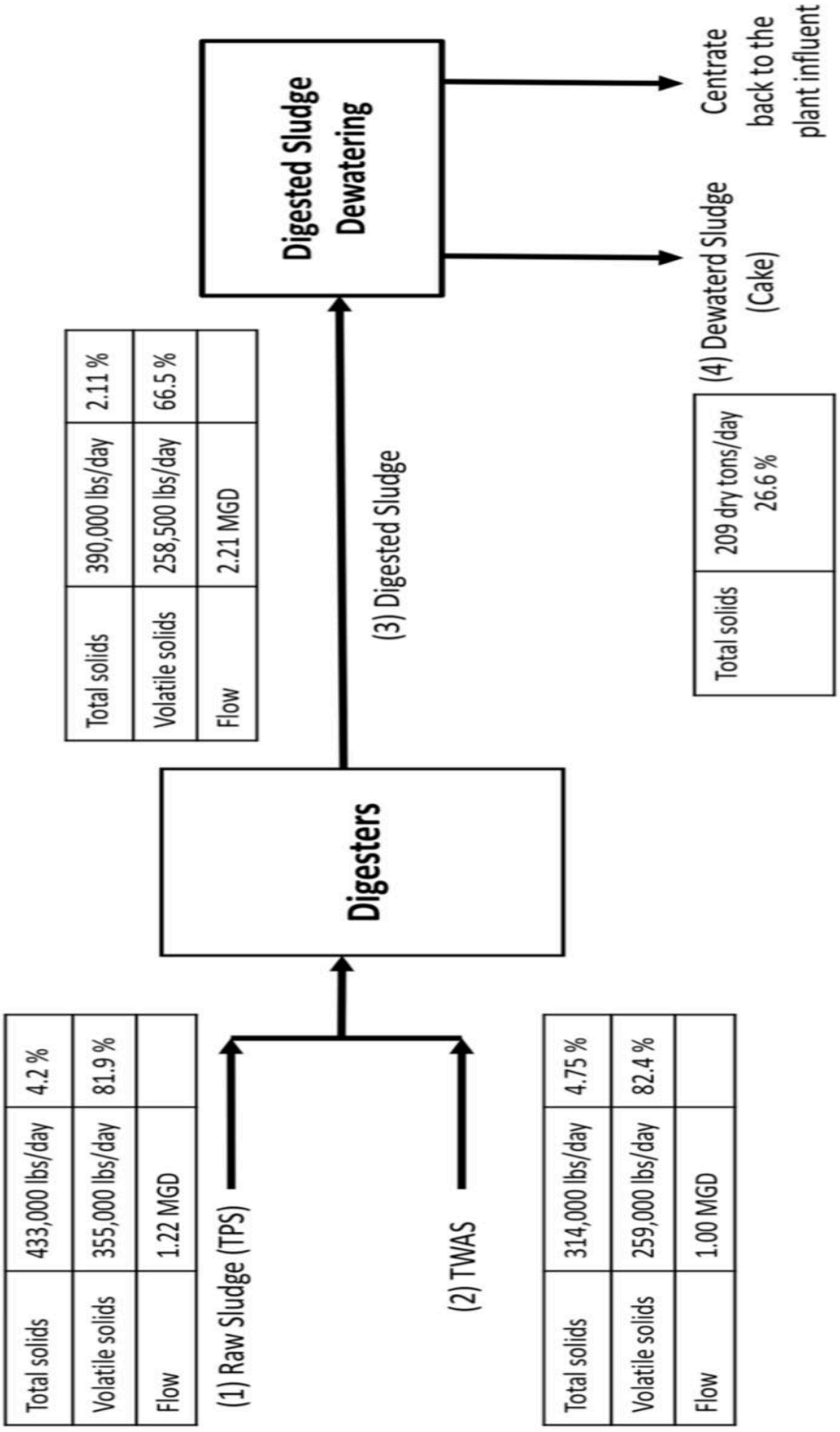


Figure 5.1. Mass and Flow rates at HWRP solid processing facility

$$\text{Non- VS}_{\text{incoming sludge}} = (\text{TS}_{\text{primary sludge}} + \text{TS}_{\text{WAS}}) - (\text{VS}_{\text{primary sludge}} + \text{VS}_{\text{WAS}})$$

$$= 133{,}400 \text{ lbs/day} \qquad \text{Eq (5.1)}$$

$$\text{Non- VS}_{\text{digested sludge}} = \text{TS}_{\text{digested sludge}} - \text{VS}_{\text{digested sludge}} = 130{,}760 \text{ lbs/day} \qquad \text{Eq(5.2)}$$

The second assumption is solid mass change over the digesters is only due to the biological destruction of VS and does not change the affinity of the radionuclides for solids or liquid. 60 percent of VS is assumed to be biodegraded in the digesters and validated from the operational data. Lastly, it is assumed that the mass of steam that is added to the digesters is negligible compared to the total liquid mass and solid mass change in the dewatering process is negligible.

Based on these assumptions, a general mass balance over the digesters and digested sludge yield from incoming sludges, the sum of TPS and thickened waste activated sludge (TWAS) is as follows:

$$\frac{solid\ mass\ digested\ sludge}{solid\ mass\ incoming\ sludge} = \frac{non\ VS\ incoming\ sludge + 0.4 \times VS\ incoming\ sludge}{mass\ incoming\ sludge} =$$

$$\frac{0.08\ mass\ incoming\ sludge + 0.82 \times 0.4 \times mass\ incoming\ sludge}{mass\ incoming\ sludge} = 0.4 \qquad \text{Eq (5.3)}$$

$$solid\ mass\ digested\ sludge = solid\ mass\ dewatered\ sludge \qquad \text{Eq(5.4)}$$

$$total\ mass\ dewatered\ sludge = \frac{solid\ mass\ digested\ sludge}{0.25} =$$

$$\frac{0.4 \times solid\ mass\ incoming\ sludge}{0.25} = 1.6 \times \text{solid mass incoming sludge} \quad \text{Eq(5.5)}$$

$$total\ mass\ incoming\ sludge = \frac{solid\ mass\ incoming\ sludge}{0.045} =$$

$$22.2 \times \text{solid mass incoming sludge} \quad \text{Eq(5.6)}$$

$$\frac{total\ mass\ dewatered\ sludge}{total\ mass\ incoming\ sludge} = \frac{1.6}{22.2} = 0.072 \quad \text{Eq(5.7)}$$

This ratio is used to calculated the counts per kilogram of sludges in Table 6.

An excel spreadsheet was created for the mass balance over the solid processing facility. Figure 5.2 is a screenshot of the excel spreadsheet.

In order to calculated the percentage of ^{131}I and ^{40}K in solids and liquid phases, and iterative process was used, a percentage was assumed, counts per kilogram of dried solids were calculated and compared to the measured values. The percentage assumption was refined until calculated activity per kilogram of dried sample was equal to the measured values. The Goal Seek function in excel was used to check the iterative process. ^{131}I and ^{40}K association with solid phase were calculated to be 13 and 37 percent respectively.

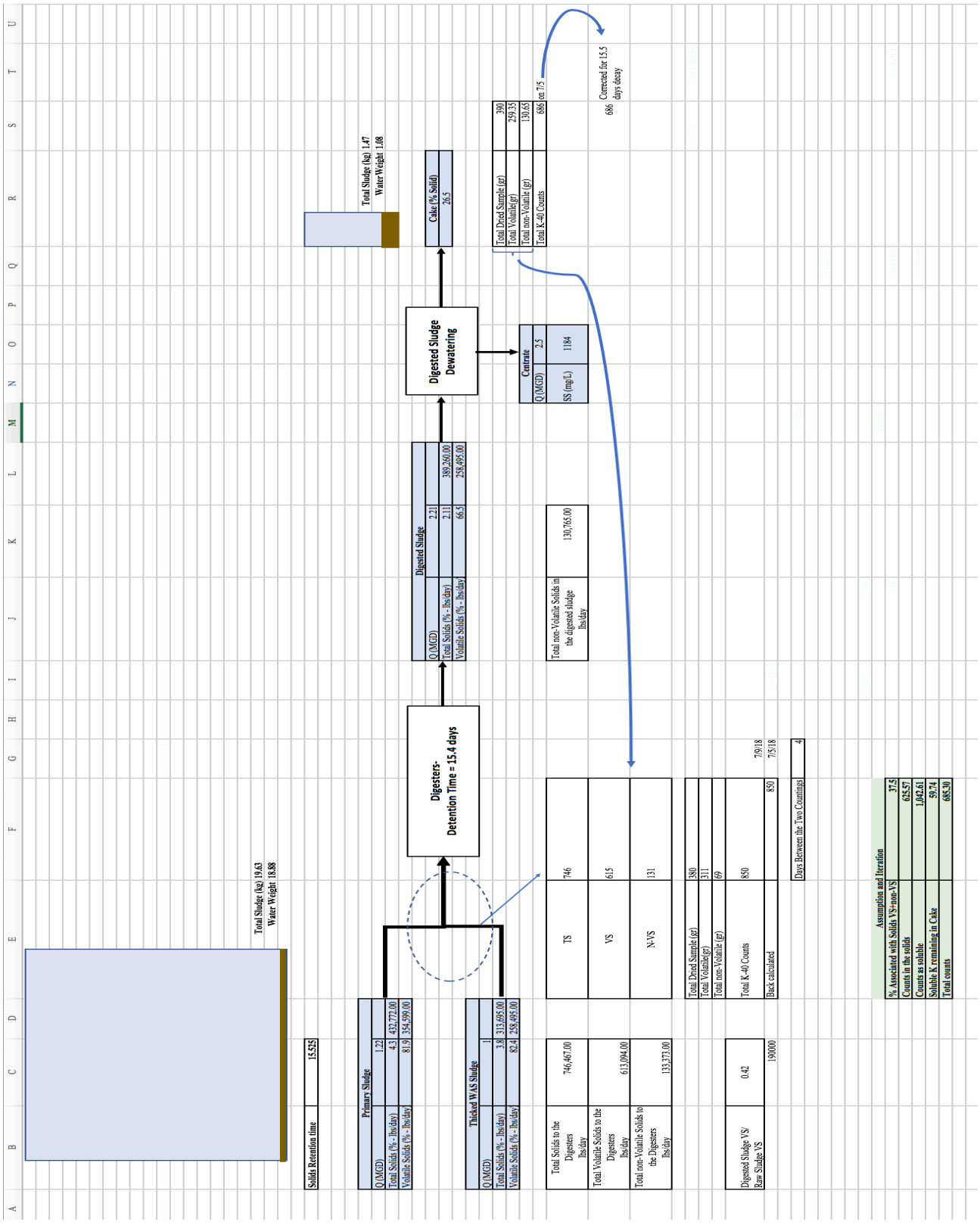


Figure 5.2. Screenshot of the excel spreadsheet developed of mass and activity balance

6. Discussion and Conclusions

All the radionuclides observed in different samples have been previously observed by others. The incinerated sludge sample is assumed to be the most concentrated form of the sample for those radionuclides that are not volatile under incineration temperatures due to minimum moisture and that the sample could be compressed even more than the ground dried sludges into the sample containers.

^{141}Ce, which was measured in the incinerated sludge sample, has been reported by (Erlandsson, 1982) down-stream of a nuclear power plant and related to this source. This radionuclide is also medically used and could potentially, like ^{131}I, be discharged to the sewer from the medical facility.

^{51}Cr, which was also measured in the incinerated sludge sample, has been reported by (Erlandsson, 1989) at concentration higher than those observed in the incinerated sludge. This radionuclide can also be discharged to the sewer from a medical facility. However, it should be noted that HWRP serves a large area of the City of Los Angeles and large volumes of sewer can dilute the concentration of source specific contaminants more than some smaller wastewater treatment facilities that serve smaller areas and process less influent flows.

^{40}K is consistently observed in all samples, which is expected due to the fact that it is coming from natural sources. It was reported in wastewater samples from different plants in the past. Similar concentration ranges are expected, since ^{40}K exists in fixed ratios in natural samples and its existence in the wastewater samples related to these natural sources, such as rocks. [7,10,15]. It

was observed in HWRP sludge samples in 1960s at levels similar, but slightly less in the raw sludge than the levels observed by the authors in the TPS and TWAS. This can be due to the fact that in 1960s, HWRP had partial secondary treatment, with no primary sludge and WAS thickening. It is shown in this paper that ^{40}K has affinity for solids and higher activities are expected in thickened sludge. The dewatered digested sludge sample is concentrated and has the highest solid percentage, therefore the highest ^{40}K in this sludge is expected compared to the other samples.

The fate of ^{137}Cs and ^{40}K and their transport in the environment is identified to correlate [10]. These are both monovalent ions, which can partially explain the similar fate. The affinity of ^{134}Cs and ^{137}Cs for solids has been demonstrated in previous studies [6], with activity ratio of the two radionuclides in dewatered digested sludge to sum of dewatered digested sludge and plant effluent ranging from 0.37 to 0.48; however, lower percentage solids association for ^{137}Cs of 10 percent has also been reported under a radiotracer study [17].

^{131}I was observed previously by many in wastewater samples, specially in plants serving medical facilities where they use this radionuclide. or downstream of nuclear power plants. These studies have reported values both higher and lower than what was measured in HWRP samples. Lower values are specially expected at HWRP compared to smaller plants that are down stream of medical facilities and have less influent flow rates that results in overall dilution of streams of contaminants.

It is apparent that during normal operation, radionuclides exist in wastewater streams at low levels, which do not cause any hazard. The annual safety limit for radiation exposure is 5 rem. (OSHA)

Rem is a unit of equivalent dose calculated from rad, which is a unit of the amount of energy absorbed in a mass of matter from radiation (0.01 J/kg) that results in ionization and release of electrons from the mass, multiplied by a quality factor (Q). The value of Q is based in the stopping power of the charged particles in water and the energy lost per unit distance travelled. Q for gamma rays has a value of 1. Therefore, rad and rem values are equal for gamma rays.

The absorbed dose of gamma rays in tissue in unit rad is nearly equal to the exposure rate, which values are typically expressed in Roentgen (R) per unit time:

Exposure rate= $\frac{\Gamma A t}{d^2}$ Eq(6.1)

Γ: Gamma exposure rate constant ($\frac{R.m^2}{hr.Ci}$) Eq(6.2)

A: Activity (Ci) Eq(6.3)

t: duration of exposure Eq(6.4)

d: Distance from the point source Eq(6.5)

Absorbed dose for tissue = 1 Roentgen ~ 1 rad Eq(6.6)

Quality factor for gamma = 1 Eq(6.7)

R gamma in tissue ~ rem Eq(6.8)

Gamma exposure rate constants are radionuclide specific. For ^{131}I and ^{40}K these values are 0.02 and 0.08 respectively $\frac{R.m^2}{hr.Ci}$ [13]. In order to estimate the absorbed dose by treatment plant operators, a hypothetical case is assumed, in which an operator spends 2 hours every day in one year (365 days) in 1 meter distance from a Primary Digester tank at HWRP, which has a volume of approximately 2.5 MG. Although the digester wall provides shielding against gamma rays, here it is assumed it to be negligible. Under this extreme hypothetical case, the operator will receive 3.2

rems in one year, which is below the safety limits. Professionals who work in facilities were there is a risk of elevated exposure to radiation are required to wear dosimeters at all time in order to make sure that the exposure safety limits are not exceeded.

However, in case of a nuclear accident or other larger atmospheric releases of radionuclides, spiked levels are expected and have been measured. [9,3,4,6,12,] The persistence of radionuclides in wastewater treatment plants after release of large amounts depends on the decay rate and affinity for solids and may take over several months to go back to pre-accident levels [12].

As extensive reuse programs are being put in place, any interruption in such programs due to contaminants could result in treats to local water supplies and very costly for metropolitans to compensate for this lost resource. Reclamation plants with reverse osmosis should protect the reclaimed water from radionuclides but will concentrate them in the brines.

Contaminated biosolids can also result in issues for reuse and recovery programs and challenging to manage. In order to take actions when spiked releases happen, it is important to know the pre-accident levels and fates of radionuclides. The results from this study serves for those purposes at HWRP. The likely fate of at least two radionuclides has been established in this study.

Many Advanced Water Treatment pilot research are in progress across Southern California and in other states. Future studies focused on fate of radionuclides in these facilities are suggested

Bibliography

1. Barquero R, Agulla M.M, Ruiz A: Liquid Discharges from the Use of Radionuclides in Medicine (Diagnosis). *Journal of Environmental Radioactivity* 2008, 99(10): 1535-1538.
2. Chino M. ,Nakayama H. , Nagai H., Terada H., Katata G., Yamazawa H: Preliminary Estimation of Release Amounts of I-131 and Cs-137 Accidentally Discharged from the Fukushima Daiichi Nuclear Power Plant into the Atmosphere. J*ournal of Nuclear Science and Technology* 2011, 48 (7): 1129-1134.
3. Durham, R.W., Joshi, S.R.: Radionuclide concentrations in two sewage treatment plants on Wester Lake Ontario, Canada, *Journal of Radioanalytical Chemistr*1979*y*, 54(1-2), 367-370
4. Erlandsson B, Mattsson S: Medically Used Radionuclides in Sewage Sludge. *Water, Air, and Soil Pollution* 1978, 9:199-206.
5. Erlandsson, B., Ingemansson, T., Mattsson, S.: Comparative studies of radionuclides from global fallout and local sources in ground level air and sewage sludge, *Water, Air, and Soil Pollution* 1982, 20, 331-346
6. Erlandsson, B., Bjurman, B., Mattsson, S.: Calculation of radionuclide ground deposition by means of measurements on sewage sludge, *Water, Air, and Soil Pollution* 1989,45, 329-344
7. Fischer, J.W., Ulbrich, S., Pittauerova, D., Hettwig, B.: Medical radioisotopes in the environment – following the pathway from patient to river sediment, *Journal of Environmental Radioactivity* 2009, 100, 1079-1085
8. Fischer, H. W., Yokoo, Y.: Preliminary comparison of radioisotope concentration in

sewage sludge after the Fukushima and Chernobyl accidents, *An open access article under the CC BY-NC-ND license* 2014 (http://creativecommons.org/licenses/by-nc-nd/3.0/).

9. Folsom, T.R. and Mohanrao, G.J. : Cesium-137 in the Effluent of a Larger City, *Nature* 1960, 4755, 979-982
10. Folsom, T. R., Mohanrao, G. J., Betz, J. M., Garber, W. F.: A study of certain radioactive isotopes in selected wastewater treatment plants, *Water Environment Federation* 1963, 35 (3), 304 – 333
11. Hoffman B.W. , Van Camerik, S.B.: A Table for Determining the True Time Representing a Count Rate Observed in Radionuclear Counting, *Analytical Chemistry* 1967, 39 (10), 1198-1199
12. Imhoff, K.R., Koppe, P., Friedrich, D. : Investigations on the concentration of radionuclides in sewage sludges of Ruhrverband wastewater treatment plants 1960 through 1986, *Water Research* 1988, 22(8), 1059-1067
13. Lauridsen, B. : Table of Exposure Rate Constants and Dose Equivalent Rate Constants**,** 1982, Downloaded from orbit.dtu.dk on: Jun 06, 2020
14. Martin, J.E. and Fenner, F.D.: Radioactivity in municipal sewage and sludge, *Public Heath Reports* 1997, 112(4), 308-316
15. Prichard M, Gesell T. F, Davis E: lodine-131 Levels in Sludge and Treated Municipal Wastewaters Near a Large Medical Complex. *American Journal of Public Health* 1981, 71(1): 47-52.
16. Rose, P.S., Swanson, R.L., Cochran, J.K.: Medically-derived ^{131}I in municipal sewage effluent, *Water Research* 2012, 46, 5663-5671

17. Stelar, E.A., Boston, H.L., Ingvar, L.L., Molbey, M.H.: Removal of radioactive cobolt, cesium and iodine in a conventional municipal wastewater treatment plant, *Water Environment Research* 1993, 65(5), 630-639
18. Van Meter, P. C., Gray J. R, Effects of uranium mining discharges on water quality in the Puerco River basin, Arizona and New Mexico, *Hydrological Sciences Journal* 1992, 37(5), 463-480
19. Yoshida N, Kanda J: Tracking the Fukushima Radionuclides. *Science* 2012, 336(6085): 1115-1116.
20. Dosimetric quantities and units (https://www.nrc.gov/docs/ML1122/ML11229A688.pdf), last accessed on 6/6/2020)
21. SOURCES AND EFFECTS OF IONIZING RADIATION, United Nations Scientific Committee on the Effects of Atomic Radiation, UNSCEAR 2000 Report to the General Assembly, with Scientific Annexes