AN ABSTRACT OF THE THESIS OF

C. Thomas Brannan, Jr. for the degree of Master of Science in Radiation Health Physics presented on June 24,1996. Title: An Investigation of Radioactively Contaminated Wastewater Reclamation Plant Biosolids.

Redacted for Privacy

Abstract approved: _____ Jack F. Higginbotham

Within the last 15 years, wastewater treatment plants have increasingly begun to find radioactive isotopes incorporated in biosolids. Regulations concerning the disposal of radioactive material via sanitary sewer systems were in place since the early 1960's and recently were further restricted. However, the updated regulations would not have prevented certain contamination cases, which may predict future incidents and additional government restrictions.

As a result, general procedures were created to investigate gross alpha, gross beta and gamma radionuclide concentrations in local wastewater reclamation plant biosolids. The US Environmental Protection Agency procedures for determining radionuclides in drinking water supplied the foundations for the tests. Supplementing the procedures, separate alpha and beta transmission curves were generated to account for detector efficiency and alpha and beta attenuation factors. The curves were designed to be effective for any gas flow proportional counter. By consulting the curves and following the procedures, 0.0 +/- 1.4 pCi/ml of gross alpha and 0.3 +/- 7.7 pCi/ml of gross beta

were measured in Corvallis Wastewater Reclamation Plant sludge. A gamma spectroscopy analysis discovered small amounts of ^{131}I and ^{7}Be at 0.11 +/- 0.01 pCi/liter and 0.96 +/- 0.08 pCi/liter, respectively. These values were in agreement with past studies.

Future government course of action regarding radioactive materials in biosolids remains uncertain, although with this study's help, Oregon State University and the State of Oregon will be prepared for future regulatory changes.

An Investigation of Radioactively Contaminated Wastewater Reclamation Plant Biosolids

by

C. Thomas Brannan, Jr.

A THESIS

submitted to

Oregon State University

in partial fulfillment of
the requirements for the
degree of

Master of Science

Completed June 24, 1996
Commencement June 1997

Master of Science thesis of C. Thomas Brannan, Jr. presented on June 24, 1996

APPROVED:

Redacted for Privacy

Major Professor, representing Radiation Health Physics

Redacted for Privacy

Head of Department of Nuclear Engineering

Redacted for Privacy

Dean of Graduate School

I understand that my thesis will become part of the permanent collection of Oregon State University libraries. My signature below authorizes release of my thesis to any reader upon request.

Redacted for Privacy

C. Thomas Brannan, Jr., Author

ACKNOWLEDGMENTS

First and foremost, I thank the Lord God for His eternal wisdom, patience and love, for without Him I can do nothing.

I would like to thank Dr. Jack F. Higginbotham for his encouragement, advice, support, and guidance throughout my MS program. He taught me to work independently and consider all of my resources. I would also like to extend my appreciation to the faculty and staff at the Radiation Center, including Dr. Brian Dodd, Dr. Kathy Higley, Dr. Steve Binney, Dr. Walter Loveland, Dave Pratt and Mike Conrady for their guidance, support, and opinions.

I would also like to thank Larry Lamperti, Mike Clark and the operators at the Corvallis Wastewater Reclamation Plant for their time and help.

Finally, I thank my wife Marla, for her constant encouragement, proven patience and undying love.

TABLE OF CONTENTS

TABLE OF CONTENTS (continued)

TABLE OF CONTENTS (continued)

LIST OF FIGURES

LIST OF TABLES

AN INVESTIGATION OF RADIOACTIVELY CONTAMINATED WASTEWATER RECLAMATION PLANT BIOSOLIDS

1. INTRODUCTION

Over 35 years ago, the Atomic Energy Commission set limits restricting the amount of radioactive material that could legally be disposed of via sanitary sewer systems. Recently as a result of contaminated biosolids (sludge) incidents, the regulations were changed restricting the material's physical form and monthly concentration levels. Even with these changes, certain contamination cases would not have been prevented. For this reason, further radioactive contamination will most likely occur resulting in the development of federal or state required periodic biosolids monitoring.

With this in mind, an investigation of radioactive material contents in the local wastewater reclamation (treatment) plant sludge ensued. An individual isotopic analysis was unnecessary considering the present stage of the regulations; therefore a general inspection of gross alpha, gross beta and gamma was performed. To correctly measure these components, original procedures were created by modifying current US Environmental Protection Agency radionuclide measurement procedures for drinking water. The varying amount of solids found in the sludge also required the generation of alpha and beta attenuation curves for finding accurate efficiencies. The purpose of this study was to create working procedures for measuring the gross alpha, beta and gamma emitting isotope concentration in sludge. Oregon State University may then use these procedures for the examination of any biosolids produced in Oregon and consequently be prepared for probable future regulations.

2. LITERATURE REVIEW AND BACKGROUND

2.1 RADIOACTIVE CONTAMINATION INCIDENTS AT WASTEWATER RECLAMATION PLANTS

A reoccurring problem involving wastewater treatment plants across the United States has been the presence of radioactive materials in wastewater biosolids. This type of contamination may become present either from naturally occurring isotopes or from sewer disposal methods from licensed facilities that utilize radioactive materials. Normally, a distinction can be made concerning the source of radioactive materials once concentrated in the sludge. The licensed isotopes used by commercial or industrial facilities usually vary widely from those found naturally. By studying the particular type of isotope, its point of origin may be determined by referring to the inventory of each nearby facility's licensed, unsealed sources.

For each of the wastewater reclamation plants (also referred to as WRP) that have experienced a radioactive contamination problem, the material originated from a licensed facility. No serious problems have occurred from natural isotopes, even though they are concentrated in sludge and treatment plant incineration ash. In the following case studies, several wastewater treatment plants that have encountered elevated levels of radioactive material in sewer lines, sludge, and incinerated sludge ash are discussed.

2.1.1 Case 1 - Tonawanda, New York

In the 1970s and early 1980s, a smoke detector manufacturer operated using ^{241}Am. Upon decommissioning the site in 1983, contaminated sewer lines leading from the facility were discovered and subsequently led investigators to find contaminated sludge and sludge incineration ash at the sewage treatment plant. In 1984, tests showed ^{241}Am levels up to 750 pCi/g in incinerator ash and 100 pCi/g in sludge. Testing of sewage reclamation plant workers revealed no radioactivity in their bones or lungs above background. (MacClennan, 1984) As a consequence of the contamination, the state of New York spent $2.5 million cleaning up the treatment plant and sewer lines and must spend an estimated additional $7 million to remediate the Tonawanda landfill. (Kennedy Jr. et al., 1992; Rimawi, 1984)

2.1.2 Case 2 - Grand Island, New York

The Tonawanda incident spurred the New York Department of Health to evaluate other wastewater reclamation plants. Dry sludge samples were taken from the Grand Island sewage treatment plant which revealed a 100 pCi/g alpha activity concentration. A nearby manufacturing facility used ^{3}H, ^{210}Po, and ^{241}Am, and subsequently discharged about 25 mCi/y. The New York Department of Labor approached the facility and requested a reduction in release concentration levels. Added filtration in the licensee's holding tank decreased the ^{241}Am concentration in sludge at the sewage treatment plant to about 40 pCi/g. Fortunately, no clean up was needed at the treatment plant. A few

workers had used the sludge as a soil supplement in their home gardens, with a measurable amount of ^{241}Am detected. However, based on sampling data, no worker received above normal doses and no extra safety precautions were implemented. (Federal Register, 1994a; GAO, 1994; Kennedy Jr. et al., 1992; Rimawi, 1984)

2.1.3 Case 3 - Oak Ridge, Tennessee

When the city of Oak Ridge put in a new WRP, contamination was found in sewage lines leading to a company specializing in the decontamination of nuclear power plant materials. Small amounts of ^{137}Cs as the primary contaminant, ^{54}Mn, ^{60}Co, and ^{134}Cs were released and had concentrated in the treatment plant sludge. The sludge had been disposed of on government owned deforested land and radiation levels at the site were 2 to 3 times background. It was later determined that the primary risk would be from consuming vegetables grown in a garden fertilized by the sludge at a dose rate of approximately 6 mrem/y. The Tennessee Division of Radiological Health set stricter release guidelines to limit the amount of radioactive material released to WRPs. Fortunately, no cleanup was needed. (Kennedy Jr. et al., 1992; Halsey, 1986; Federal Register, 1994a)

2.1.4 Case 4 - Royersford, Pennsylvania

A commercial laundry for radioactively contaminated clothing was discharging approximately 15,000 gallons of wastewater to the local treatment plant per day. An

inspection in 1985 revealed radiation levels up to 1.2 mR/h at the secondary digester. The Nuclear Regulatory Commission (NRC) inspections showed the licensee had complied with the regulations. Temporary holding of the wastewater, treatment to adjust the pH, and gross alpha and beta activity analyses proved federal laws had not been violated. Still, increased concentrations were found in farmer's fields from sludge applications. The NRC evaluated the impacts finding the highest potential doses would be received by farmers working in their fields, estimating a value less than 5 mrem/y. Radiation measurements taken on the outside of a sewage tanker truck ranged up to 0.3 mR/h, well within the Department of Transportation limits. Like the other cases no remediation efforts were needed for this situation. (Federal Register, 1994a; GAO, 1994; Kennedy Jr. et al., 1992; NRC, 1986a)

2.1.5 Case 5 - Erwin, Tennessee

The Erwin WRP must spend an estimated $250,000 to clean up a sludge digester. Nuclear Fuel Services (NFS), the sole licensed radioactive material user of the Erwin treatment plant, had legally discharged small amounts of ^{241}Am, ^{239}Pu, ^{232}Th, ^{234}U, ^{235}U, and ^{238}U via the sanitary sewer system. The contamination, found in 1986, prompted NFS to reduce possible waste streams including laundry and laboratory drains. Proportional samplers were also installed to monitor flow rates and collect samples. (GAO, 1994)

2.1.6 Case 6 - Washington, DC

Several federal government facilities with a broad spectrum of radionuclides discharge to the Blue Plains Wastewater Treatment Plant in Washington, DC. Part of the waste goes directly into the sanitary sewer while a portion is retained in holdup tanks to allow the short-lived nuclides to decay before being discharged. Two NRC inspections in early 1986 found no violations at the facilities, although 27 different radionuclides have been found at the treatment plant. Samples taken from facility effluents contained 2% or less of the specified limits for maximum daily release concentrations as stated in 10 CFR 20. Sewage treatment plant analyses showed concentrations of ^{137}Cs and general beta emitters were on the same order of magnitude for liquid influent and effluent, with sludge concentrations about 10% of the liquid effluent values. However, influent concentrations of alpha-emitters were measured to be 10 times higher than liquid effluent concentrations. Despite these findings, no actions were necessary for cleanup. (Kennedy Jr. et al., 1992; NRC, 1986b; Federal Register, 1994a)

2.1.7 Case 7 - Portland, Oregon

In 1989, the city of Portland and the State's Health Division mandated that a state licensee clean up sewer lines and install a pretreatment system. As a result of disposing ^{232}Th as thorium oxide to the sewage treatment plant, the company had to pay approximately $2 million to remedy the situation. City employees are currently required to wear protective clothing when working in sewer lines that contain thorium oxide

sediment. State and local authorities evaluated a solution to completely stop the discharges from the licensee. However, the City of Portland was uncertain whether it could legally require the licensee to discontinue the thorium oxide discharges, which would cost approximately $5 million according to the licensee. The City decided the case lacked clear, scientific standards and requiring a discontinuation order of the licensee's discharges would not hold up in court. (GAO, 1994)

2.1.8 Case 8 - Ann Arbor, Michigan

In Ann Arbor during 1991, ^{60}Co, ^{54}Mn, ^{110m}Ag, ^{108m}Ag, and ^{65}Zn contamination were found although, fortunately, no remediation or clean up was required at the WRP. (GAO, 1994)

2.1.9 Case 9 - Cleveland, Ohio

Created in 1972, the Northeast Ohio Regional Sewer District, NEORSD, is composed of four sewage treatment plants: Easterly, Southerly, Strongville, and Westerly. The Southerly plant must handle and properly dispose of sludge from each plant excluding Westerly. Located in Cuyahoga Heights, the Southerly plant can treat up to 175 million gallons per day and serves over 500,000 Cleveland and suburban area residents. Southerly is one of the largest activated sludge treatment plants in the nation.

The combined sludge from the treatment plants is either incinerated or taken off site to a district approved landfill. The incinerated sludge is pumped as a slurry and placed

in three ponds allowing it to settle and evaporate. Once the ponds become full, the ash is moved to various fill locations on site.

By shear coincidence in April 1991, an NRC aerial radiological survey investigating a nearby former licensee, happened to detect elevated levels of radiation at the Southerly plant. On May 15, 1991, two Ohio Department of Health officials and an NRC radiation specialist conducted surveys and obtained soil samples in order to more precisely determine the location of the ground contamination. Radiation monitoring detected levels at about 20 times background in the northeast area while collected samples contained 27 to 79 pCi/g of ^{60}Co. ^{226}Ra and ^{137}Cs were also detected but were at expected naturally occurring levels. Further surveys by Oak Ridge Associated Universities (ORAU) indicated exposure rates from 15 to 580 μR/hr. The ^{60}Co limit for areas released for unrestricted use as determined by the NRC is 8 pCi/g. In contrast, the maximum surface soil concentration found was over 3×10^6 pCi/g. ORAU did not consider this sample representative of the area soil concentrations.

Even though above normal radiation levels were detected, the NRC stated that no indicator was found to cause significant radiation exposure to the public because of the fixed location of the contamination. Whole body testing of Southerly workers also found no radiation levels above normal, which was expected considering the half life of ^{60}Co and timely elimination rates from the body.

As of mid-February 1994, the district had spent $900,000 for on site remediation activities and an additional $120,000 for fencing to prevent public access. Thus far, the

district is planning to keep the ash on site and has already placed 174,000 cubic yards in a fill area with six inches of clean soil as a cover.

Advanced Medical Systems, Inc. (AMS), a radioactive source manufacturing plant, was the cause of these difficulties. AMS contended they had not exceeded the NRC's limits for sewage disposal of ^{60}Co and the NRC agreed. Inspections found no violations and the manufacturer's records showed a total of 0.2 Ci of ^{60}Co was discharged from May 1980 to May 1989. According to NEORSD's lawyer, Tom Lenhert, only 65 mCi should have been present at Southerly after allowing for decay, when in fact, Southerly has over a half a curie in its inventory - a factor of 10 greater. (Nuclear News, 1994; GAO, 1994)

As a result, the district filed two petitions to the NRC. One on March 3, 1993, requested modifications in the AMS NRC license to (1) assume all costs that resulted from the ^{60}Co releases to the Southerly plant and (2) decontaminate the sewer lines that connected the manufacturer to the treatment plant and (3) continue to decontaminate downstream sewers for as long as necessary. Then on August 3, 1993, the district issued another petition that required the manufacturer to provide adequate financial assurance to cover public liability. Also separately stated was a petition for rulemaking that asked the NRC to amend the regulations to (1) require licensees to provide at least 24 hours advance notice to the appropriate sewage plant before releasing radioactive material to the sanitary system and (2) exempt sanitary waste stream materials from the NRC approval requirements for incineration. Consequently, an advanced notice of proposed rulemaking was issued in the Federal Register on February 24, 1994, by the NRC that asked for comments and information with regard to the need for an amendment concerning

radionuclide release into sanitary sewers. The comment period ended May 26, 1994 with no word of possible changes or outcomes. (Federal Register, 1994a)

2.2 WASTEWATER RECLAMATION PLANT (WRP) CHARACTERISTICS

The discharge of a wide variety of suspended solids, dissolved solids, organics, inorganics, chemicals, grease, biological oxygen demand, gases and other materials into a collection system from domestic and industrial sources commonly occurs. The predominant means of transporting these types of waste is from the water flow. Whether they be from natural sources, such as rain and melting snow, or man-made systems, pollutants are localized as water converges on rivers and lakes creating the need for treatment. Wastewater treatment systems became a necessity as populations grew, and then became law in the early 1960s in order to protect people and the environment. Today, numerous configurations of waste water treatment facilities work to remove contaminants from the influent before discharging the resulting effluent. As a result, large amounts of sludge are generated. By the year 2000, it is estimated that about 12 million metric tons (dry weight) of municipal sludge will be generated each year. (Kennedy Jr. et al., 1992)

Notably, a wide variety of system arrangements, hardware and treatment methods are in use, with few facilities designed exactly alike. The purpose of this chapter is not to delve into the details of wastewater reclamation methods, but rather to expand awareness of general treatment processes and equipment. Many fine references may be consulted for

further details and information regarding wastewater treatment techniques that are not covered here. (Metcalf and Eddy, 1991; Tchobanoglous, 1987; Wheatley, 1990)

2.2.1 Methods of Treatment

Several factors determine the treatment methods for each particular plant. They include population size, industrial impacts, influent content, effluent release point, and discharge requirements. Obviously, larger volumes of wastewater require larger facilities, which are directly related to the number of people and number and type of industrial complexes that utilize the treatment plant.

Most modern water reclamation facilities in use today incorporate physical, chemical, and biological processes into their systems. However, it may be difficult to distinguish each individual treatment method separately because they often overlap throughout the system.

Physical processes involve the use of physical forces in order to rid the influent from particulate and soluble materials. Generally, the first physical process wastewater encounters is a screen or metal bars spaced closely together. Screening removes large debris such as sticks, shoes, and cloth which could potentially damage pumps and other systems. Depending on the plant, the debris is removed from the screen either automatically or manually. Grit, pebbles, or small rocks are eliminated from the waste stream using sedimentation. Sedimentation is used in various stages where heavier matter settles to the bottom of tanks and can then be removed. Comminution is another physical process which cuts the remaining solid objects into small pieces to protect any piping or

valves from damage. During aeration, air passes through waste water reducing the volatile gas content in the water such as hydrogen sulfide, essentially removing taste-causing and odor-causing substances. (Tchobanoglous, 1987) Mixing, adsorption, and filtration also contribute to the removal of particulate and soluble materials. Other methods include flocculation, an aggregation of particles, and reverse osmosis, the forcing of higher concentration levels to lower concentration levels through a membrane.

Chemical methods use chemicals or chemical reactions to clean the water. Examples of these techniques include disinfection by chlorine gas or other chemicals to remove pathogenic organisms, chemical precipitation which generally removes metal ions, and coagulation which destabilizes colloidal particles so that particle growth occurs during flocculation. Other chemical methods also include oxidation and ion exchange which primarily remove undesirable ions such as Fe^{+2} and Mg^{+2}.

Biological methods use biological means for the removal of bacteria, organisms, nitrogen and phosphorous among others. Biological processes typically incorporate suspended or attached growth (depending on location of the microorganism) and an aerobic or anaerobic (depending on metabolic activity) activated sludge process. Using microbial cultures, bacteria and other organisms are broken down, or metabolized under controlled systems. However, as a result of this process, further microbial cultures are produced. If a method of wasting these cells is not used, system failure occurs. Therefore plant operators must keep the system balanced by altering temperatures and flow rates which allows the microbial mass to remove bacteria and grow while concurrently removing the excess cultures.

Activated sludge processes are also being increasingly used for the removal of the inorganic nutrients like nitrogen and phosphorous. (Tchobanoglous, 1987) Trickling filters are an example of aerobic attached growth treatment that allows the influent to pass over objects such as 2-10 cm diameter rocks or plastic material. As the influent is sprayed on top of the permeable medium, microorganisms grow and generally remove organics and perform nitrification allowing for denitrification to occur later in the process. Rotator biological contactors use the same principles as the trickling filter, although the permeable media such as polyethylene rotates on a shaft while partially submerged.

2.2.2 Treatment Stages

The methods of treatment discussed previously may be incorporated at WRPs in varying stages. These stages include primary, secondary, and tertiary treatment. Until the 1972 Federal Water Pollution Control Act, primary treatment was the only level of treatment practiced at most municipal treatment plants. (Ainsworth, 1994) Primary treatment typically involves screening, grit removal, flow measurement, comminution, and sedimentation through primary settling. These physical methods remove the suspended organic solids and floating materials at efficiencies between 33 to 56 percent; however the levels of bacteria and pathogens remain high. (Lester, 1987)

The Federal Water Pollution Control Act required the addition of secondary treatment to WRPs. Secondary treatments employ biological processes to break down sludge generated from primary treatment and remove organic compounds, nitrogen and phosphorous. Some secondary processes include, but are not limited to, biologically

activated sludge, extended aeration, trickling filters, aerated lagoons, and anaerobic digestion. Effluents leaving this stage of treatment generally have a low biochemical oxygen demand (the amount of oxygen needed to metabolize biodegradable organics) and little suspended solids.

Tertiary treatments are used less extensively but are sometimes needed to conform to regulatory effluent requirements. Processes involved here can also be used in primary or secondary treatments steps. When downstream of secondary treatment, they are considered tertiary. Examples of tertiary processes include filtration, nitrification, microscreening, ion exchange, and reverse osmosis. (Tchobanoglous, 1987)

The sludge itself undergoes changes through various techniques before the final disposal form is reached. These include thickening by centrifugation or dewatering with lagoons that greatly reduce the volume, stabilization through digestion and composting and incineration using pyrolysis or starved-air combustion. (Ainsworth, 1994)

2.2.3 Sludge Discarding and Removal Techniques

The generation of sludge occurs strictly as a result of complying with regulations concerning effluent discharge. Sludge, which contains byproduct materials, has both liquid and solid components with the solids portion generally not exceeding 10%. Primary settling, sedimentation, and secondary biological treatments are responsible for generating the bulk of the sludge residue. Most sanitary engineers agree that the hardest part of implementing a WRP is the disposal of the removed material.

Plant size plays a major role in the determination of the sludge disposal options for each particular plant. Small treatment plants which produce small amounts of sludge use lagoons and drying beds for temporary storage. Sludge is then buried in a landfill or converted to fertilizer for land application. For intermediate to large WRPs, sludge is thickened before digestion and dewatered. Sludge incineration may then be utilized to further decrease the overall waste volume (by approximately 95%) because disposal space is usually limited. Most often the end waste product is trucked away to landfills or for land spreading. (Kennedy Jr. et al., 1992)

Land applications of sludge are used to provide nutrients and condition soils which result in healthier, increased growth. The most common application practice utilizes tank trucks that spray liquid sludge evenly across the surface. However, other practices exist which include the application of dried sludge using a distribution machine or wet sludge by injection into the topsoil. Areas of application that benefit from the sludge's unique properties include agricultural fields, non-agricultural sites such as public parks, forests, and disturbed land. Each area has specific loading characteristics with individual sludge concentrations varying for each recipient. Precautions must also be taken when sludge is to be disposed of at designated disposal sites or landfills. Whether the disposal site was designed exclusively for sludge, or in conjunction with other solid wastes, certain considerations cannot be forgotten such as limiting public access, testing groundwater seepage or runoff, and evaluating potential hazards.

2.3 SEWER AND EFFLUENT RADIONUCLIDE REGULATIONS

Often, federal agency's jurisdictions overlap within industries. Such is the case with waste water treatment plants. The Nuclear Regulatory Commission (NRC) and the Environmental Protection Agency (EPA) each control certain aspects regarding sewer system releases, treatment plant operations, and solid, liquid and gaseous effluents. Ironically, no regulations from any agency exist regarding the determination of sewage sludge radioactivity detection limits or maximum concentrations of radioactive material. This section discusses the current regulations regarding sanitary sewer issues and distinguishes between each regulatory body's concerns regarding radioactive sewage sludge.

2.3.1 General WRP Regulations (EPA)

The EPA has the most knowledge and control over general WRP procedures and practices. The Agency is responsible for overseeing the National Pretreatment Program under the Federal Water Pollution Control Act, commonly known as the Clean Water Act. According to 40 CFR 257, industrial waste dischargers must comply with national pretreatment standards that protect treatment plants, workers, and sewage sludge from pollutants. The EPA also regulates the use and disposal of sewage sludge under this act. Sludge incinerator ash is controlled under the Resource Conservation and Recovery Act (RCRA) of 1976, as amended.

With all these EPA regulations affecting sewage influent and effluent, the EPA cannot control radioactive materials included in the Atomic Energy Act (AEA). These are in the Nuclear Regulatory Commission's domain. Strangely enough, the EPA is allowed to regulate naturally occurring radioactive materials (NORM) which sometimes concentrate in WRP sludge. Also, under the Atomic Energy Act of 1954, as amended, and the Reorganization Plan No. 3 of 1970, the EPA may establish generally applicable environmental standards as long as they are for the protection of the environment at large. The Agency, therefore, under the Clean Air Act has the authority to regulate air emissions from incinerated sewage sludge that may contain AEA radioactive materials. In a round about way, National Emission Standards for Hazardous Air Pollutants (including radionuclides) may indirectly control the concentrations of radionuclides in sewage sludge and ash. This remains to be seen.

Methods have been developed for the measurement of gross alpha, beta and gamma activities in drinking water with limits that must comply with the Safe Drinking Water Act, PL 93-523, 40 FR 34324. These methods are contained in *Prescribed Procedures for Radioactivity in Drinking Water* written by Herman L. Krieger and Earl L. Whittaker in 1980. Procedures included have the ability to determine gross alpha, gross beta, gross radium, gamma emitting radionuclides and, in particular, radioactive cesium, strontium, iodine, tritium and uranium. (Krieger and Whittaker, 1980)

The National Interim Primary Drinking Water Regulations (NIPDWR) promulgated by the EPA require a gross alpha detection limit of 3 pCi/l for 40 CFR 141.15(a) and a gross beta detection limit of 4 pCi/l. (Krieger et al., 1980) When testing

surface or ground waters, if the gross alpha activity exceeds 5 pCi/l, the same or an equivalent sample must be analyzed for alpha-emitting radium isotopes and when a gross beta analysis exceeds 15 pCi/l a sample must be tested for ^{89}Sr and ^{134}Cs. Should a gross beta sample exceed 50 pCi/l in surface and ground waters, the identity of the major radioactive constituents must be found and appropriate organ and total body doses determined. (EPA, 1986; 40 CFR 141, 1995)

As expected, the EPA also regulates the amount of gamma emitting radionuclides in drinking, surface and ground waters. The limits set forth in PL 93-523, 40 FR 34324 recommend that, in the case of man-made radionuclides, the limiting concentration is that which will produce an annual dose equivalent to 4 mrem/yr. This limit is found by using the 2 liter per day drinking water intake using NBS Handbook 69. (Krieger and Whittaker, 1980; 40 CFR 141, 1995)

In 1988, the EPA performed a national sewage sludge survey but decided not to look for radionuclides because a literature review showed the problem was not widespread. It was assumed that radionuclide levels would not be of concern. However, Alan Hais of the EPA notes in a *Science News* article that cost concerns overrode any thoughts about investigating radionuclides. (Marino, 1994) On the other hand, in 1994 EPA was designing a sewage sludge study for 1996 that would look for ^{131}I, ^{226}Ra, ^{241}Am, and ^{137}Cs. If monitoring sludge eventually becomes the EPA's responsibility, Hais maintains that the process is so complicated, any changes would not take effect until the year 2000, unless EPA finds a need to accelerate rulemaking. (Marino, 1994)

On March 6, 1996, staff of the Division of Waste Management and Office of Research attended a briefing by Ramona Trovato, Director of the EPA Office of Radiation and Indoor Air. Other representatives from the Water Environmental Federation and several Publicly Owned Treatment Works (POTW) also participated in the briefing hosted by the Association of Metropolitan Sewerage Agencies (AMSA). AMSA reported preliminary results from a radionuclide concentration in sludge and ash survey participated in by 50 wastewater plants in at least 18 states. Thus far, results suggest that the naturally occurring potassium and radium are present in significant quantities, but the survey was limited. Industry representatives discussed the possibility of a jointly funded NRC/EPA extensive sludge and incinerator ash survey which would further evaluate the need for NRC rulemaking. They also explained their continued interest in a joint guidance document addressing radioactive material reconcentration at WRPs. Subsequent meetings to discuss this issue and the NRC/EPA survey are planned. (Weekly Information Report, 1996)

2.3.2 Sewer Release Regulations (NRC)

The NRC has no jurisdiction regarding sanitary sewer systems, sludge contents, and sludge disposal. In contrast, the NRC governs the release of radioactive materials into the sanitary sewer and prescribes regulations (which are always subject to change) that determine release limits, located in 10 CFR 20.2003 entitled *Disposal by Release Into Sanitary Sewerage*. The regulations state that a licensee may dispose of radioactive materials as long as the material is readily soluble (or readily dispersible biological

material) and the quantity does not exceed the value listed in table 3 of appendix B to §§20.1001-20.2401. This value is the average monthly concentration in the total volume of sewage released by the licensee. The concentration values were derived by taking the most restrictive occupational oral ingestion annual limit of intake (ALI) and dividing by 7.3×10^6 (ml). This factor is composed of the annual water intake by ICRP's "Reference Man" of 7.3×10^5 (ml) and a safety factor of 10. These concentrations, if the sewage released by the licensee were the only source of water ingested by a reference man throughout a year, would result in a committed effective dose equivalent of 0.5 rem (combining the 5 rem worker occupational limit with the 0.1 rem annual dose limit for the public).

The total quantity of radioactive material that can be released to the sanitary sewer can not exceed 5 Ci/y (185 GBq) of ^3H, 1 Ci/y (37 GBq) of ^{14}C, and 1 Ci/y (37 GBq) of all other radioactive materials combined. Excreta from hospital patients undergoing diagnosis or therapy using radioactive materials is exempt. See Appendix 1. for the full text of 10 CFR 20.2003 and Appendix 2. for selected individual isotope concentration limits. (10 CFR 20, 1995)

2.3.2.1 A History of NRC Sewer Release Regulations

Over 35 years ago, the Atomic Energy Commission established a basis for radioactive material release into sanitary sewers with few changes until 1986, when the NRC proposed significant modifications in the provisions for release of radioactive material into sanitary sewerage as stated in 10 CFR 20.303. Because of the possibility of

multiple contributors, the dilution in the system could not be counted on to achieve acceptable effluent concentrations. The NRC proposed an average concentration limit in the total volume of sewage released in a month based on the 0.5 rem committed effective dose equivalent to reference man. The concern was over public exposure downstream from sewage plant liquid effluent release points, and did not consider sludge reconcentration. (Federal Register, 1986) As case histories show, the problems continued.

In 1991, the NRC made changes restricting the disposal of nonbiological insoluble materials for the first time in 30 years. Originally "dispersible wastes" mentioned in the proposed rules would have been disallowed; however a large number of commentors felt that allowing only soluble waste would have an adverse impact. Those who grind up animal carcasses would have no longer been able to dispose of the ground residue in the sanitary sewer. The NRC changed the proposed rules to allow "dispersible biological materials", in addition to radionuclides in soluble form so long as the average monthly concentration limits were met. Also in 1991, because of past contamination incidents (mainly from ^{60}Co and ^{241}Am) and the change from 500 mrem to 100 mrem in public dose limits, the concentration limits were reduced by a factor of 10, but the yearly release limits remained unchanged. Licensees had to comply by January 1, 1994. (Federal Register, 1991)

With the regulations recently modified regarding radioactive material release into the sewer, an unexpected event occurred. The Northeast Ohio Regional Sewer District (District) discovered contaminated sludge on site. On August 10, 1993, the NRC docketed a petition for rulemaking (Docket No. PRM-20-22) from the District asking for

a change in regulations. They requested an amendment to the current regulations requiring all licensees to provide at least 24 hours advance notice to the appropriate sewage treatment plant before releasing radioactive material to the sanitary sewer system. The District also requested a change in the regulations which prohibit the incineration of radioactive waste without NRC approval, to exempt materials that enter the sanitary stream under 10 CFR 20.2003. (Federal Register, 1993)

The February 24, 1994 Federal Register contained an advanced notice of proposed rulemaking asking for comments on further revisions of the regulations pertaining to discovery of radioactive materials in WRPs. In particular, the NRC requested information and comments regarding possible changes in the form of the disposal material, the total quantity of material, type of limits, and patient excreta exemption. They questioned whether or not the number of licensees releasing to the same treatment plant, or plant size and systems should be considered for regulatory changes. A possibility existed that, by incorporating current sewer treatment technologies, the contemplated rulemaking would improve the control of radioactive materials disposed of in the sanitary sewer. (Agenda of Regulations, 1996) The Commission also speculated whether or not the use of a dose limit approach for concentration limitations is necessary. This would involve writing total quantity and concentration values in a Regulatory Guide to facilitate compliance with the dose limit. The comment period ended May 26, 1994, and currently the publish date of the proposed rules is undetermined. (Federal Register, 1994a)

Alternatively, in December 1994 the NRC published a notice of availability of a generic dose assessment for disposal of incinerator ash in a landfill. They concluded that

disposal of incinerator ash at Appendix B, 10 CFR 20 concentrations was acceptable for most radionuclides. Comments regarding the generic dose assessment expired February 13, 1995. (Zeyher, 1995, from Federal Register, 1994b)

2.3.2.2 Studies Aiding the Development of Regulations

To aid the determination of radioactive concentration limits, the NRC hired Pacific Northwest Laboratory in Richland, Washington to perform pathway exposure analyses. The study considered eleven scenarios with parameters that provided conservative results, rather than worst case results. It concluded that five critical radionuclides have the potential to cause doses in excess of 10 mrem/y if licensees discharged quantities nearing the 1 Ci/y limit. These radionuclides were ^{60}Co, ^{90}Sr, ^{137}Cs, ^{192}Ir, and ^{241}Am. From the eleven scenarios, the Sludge Process Operator scenario had the potential to receive the greatest dose, 360 mrem/y from ^{60}Co. (Other scenarios and outcomes can be found in the report.) Sensitivity analyses were also performed, noting the inventory of radioactive materials in a sanitary sewer system to be the most sensitive parameter. The next most sensitive parameters were river flow rate, radioactive decay time, and χ/Q.

The study did not include excreta from those patients receiving radioactive therapeutic and diagnostic treatments. A separate study considering potential doses from this factor will be organized in the future. Finally, the study concluded that the disposal of radioactive materials using the sanitary sewer may not be insignificant and needs further research. (Kennedy, Jr. et al., 1992)

2.3.2.3 Medical Isotopes

The regulation of radioactive patient excreta has instigated an ongoing discussion since its inception. The argument that hospitals should comply with the same regulations as other licensees is not new. Nuclear power and other commercial industries feel the NRC regulations are not consistent because of hospital exemptions and should either be less restrictive for commercial restrictions or more restrictive regarding patient excreta. Today, radioactive disposal of human excreta remains unregulated for several reasons.

- Sanitary sewer systems are specifically designed to control human excreta.
- Radiation protection methods are maximized by eliminating handling and storage when the sanitary sewer is used.
- Other health considerations for handling human excreta exist beyond exposure concerns.

2.3.3 Licensed Facilities in Corvallis

Because Oregon is an NRC agreement state, it oversees the licensing and regulation compliance procedures. Each individual entity in Oregon must obtain a license for the possession and use of AEA radioactive materials. The City of Corvallis has several groups that are licensed and commonly utilize a wide variety of radioactive substances.

The first licensee is a company called Antivirals, Inc. Located in the southern part of Corvallis, Antivirals, Inc. is licensed to use ^{32}P in aqueous nucleotide triphosphate form and may not possess more than 25 mCi at any one time. The company is also licensed to

use ^{35}S in amino acid aqueous form with a maximum quantity of 50 mCi for genetic research.

The second largest user of isotopes with sanitary waste treated by the CWRP is Good Samaritan Hospital with a priority 3 medical license. As shown in Table 1., a wide variety of radionuclides are used for uptake, dilution and excretion studies, brachytherapy, and diagnostic and therapeutic uses. Table 1. is not meant to be all inclusive, but to provide a general listing of the most frequently used isotopes at this particular hospital.

Oregon State University (OSU) is the largest user of radioactive materials in Corvallis. The broad scope type A license allows OSU to possess up to 120 curies (1500 mCi each, with exceptions) of any radioactive material between atomic numbers 1 and 83 and up to 115 mCi (5 mCi each, with exceptions) of any radioactive material with atomic numbers between 84 and 103 in any physical or chemical form. The license also takes into account the use of OSU's 1 MW TRIGA reactor, licensing any radioactive material with a half-life of 24 hours or less, incident from the irradiation of samples in the reactor. Most of the radionuclides are used for research and development, instrument calibration, and moisture and density measurements, among others things.

As previously discussed, according to 10 CFR 20.2003 each of the licensed users of radioactive materials may legally dispose of up to 1 Ci/y of ^{3}H, 5 Ci/y of ^{14}C, and 1 Ci/y of all other isotopes combined into the sanitary sewer, notwithstanding the fact that Good Samaritan Hospital patient's radioactive excreta are exempt. Even though the regulations permit such disposal limits, the licensees release extremely small fractions of the legal

limit. Oregon State University, having the largest quantity and widest variety of radionuclides in Corvallis, strictly enforces its no release policy.

Radionuclide[1]	Role[2]
^{67}Ga	Tumor and inflammatory lesion imaging
^{89}Sr	Bone cancer pain relief
^{99m}Tc	Brain, heart, lung, thyroid, gall bladder, skin, lymph node, bone, liver, spleen, and kidney imaging
^{111}In	Abdominal infection imaging Tumor localization Brain and kidney imaging Cerebrospinal fluid labeling Metastatic melanoma imaging
^{123}I	Blood pool imaging Brain, lung and renal circulatory system scanning Reticuloendothelial system imaging
^{125}I	Osteoporosis detection Tracer for drugs Prostate and brain cancer treatment
^{131}I	Thyroid disorders Brain biochemistry in mental illness Treating b-cell lymphoma (Monoclonal Antibody) Lymphoid tissue tumor, hyperthyroidism
^{133}Xe	Lung ventilation studies Regional cerebral blood flow studies Liver imaging

Table 1. Commonly Used Isotopes at Good Samaritan Hospital for Therapeutic and Diagnostic Purposes

[1](Radiology Department, Good Samaritan Hospital)

[2](Binney, 1995)

2.4 THE CORVALLIS WASTEWATER RECLAMATION PLANT

Now that general wastewater treatment plant configurations and regulations binding plant procedures have been discussed, it is possible to go into more detail regarding the City of Corvallis' Municipal Wastewater Reclamation Plant and upcoming remediation activities.

The City of Corvallis, Oregon, has one wastewater treatment plant serving approximately 47,480 residents, according to the Corvallis City Planner. Operators are on duty 24 hours a day monitoring systems and performing most maintenance duties. The public is always welcome for informative tours of the facilities during the normal work week.

2.4.1 History

The Corvallis Wastewater Reclamation Plant (CWRP) was first constructed in 1952. Before the plant existed, the City of Corvallis like most other riverside communities discharged untreated sewage directly into the Willamette River. The initial plant only used a primary treatment system consisting of comminution, grit removal and a clarifier, but this was the beginning of a move to stop pollution release into the river. In 1965, the plant was expanded to include trickling filters as part of the secondary treatment facilities. An increase in sewage flow from the growing City of Corvallis, in conjunction with stricter discharge regulations from regulatory agencies, led to the development of a long range plan in 1973. The plan was designed to handle the city's wastewater treatment

requirements in three separate stages through the year 2005 utilizing expansion and improvements. Each phase of the plan was to take place in 10 year intervals or as actual population growth dictated. (*City of Corvallis*, 1978)

Under the guidance of the consulting and engineering firm Brown and Caldwell, Contractors, Inc., construction of the first stage began in 1976 under Mayor Donald L. Walker (1975-1978) in order to expand existing facilities to accommodate about 10 million gallons per day. An activated sludge system was also designed and built in order to comply with new effluent regulations. At the completion of the first stage in June 1978, the treatment plant was able to serve a population of 64,000 and the total cost of the project was $9,193,000 (funded by EPA grants and general obligation bonds). The second and third stages were designed to accommodate equivalent populations of 84,000 and 103,000, respectively. However, Corvallis population growth projections were inaccurate and the second and third stages proved unnecessary. Thus far, no further plant improvements have been made, although the city council recently approved a combined sewer overflow remediation plan which includes modifications (discussed in section 2.4.1.3 Corvallis' Combined Sewer Overflow Status).

2.4.2 Wastewater Treatment Methods and Layout

The Corvallis Wastewater Reclamation Plant utilizes primary and secondary treatment processes. Compared to numerous other treatment facilities, it is typical for a city of equal size. Figure 1. shows the layout of plant systems and influent and effluent paths which will aid in the discussion. Raw sewage enters the plant through two large

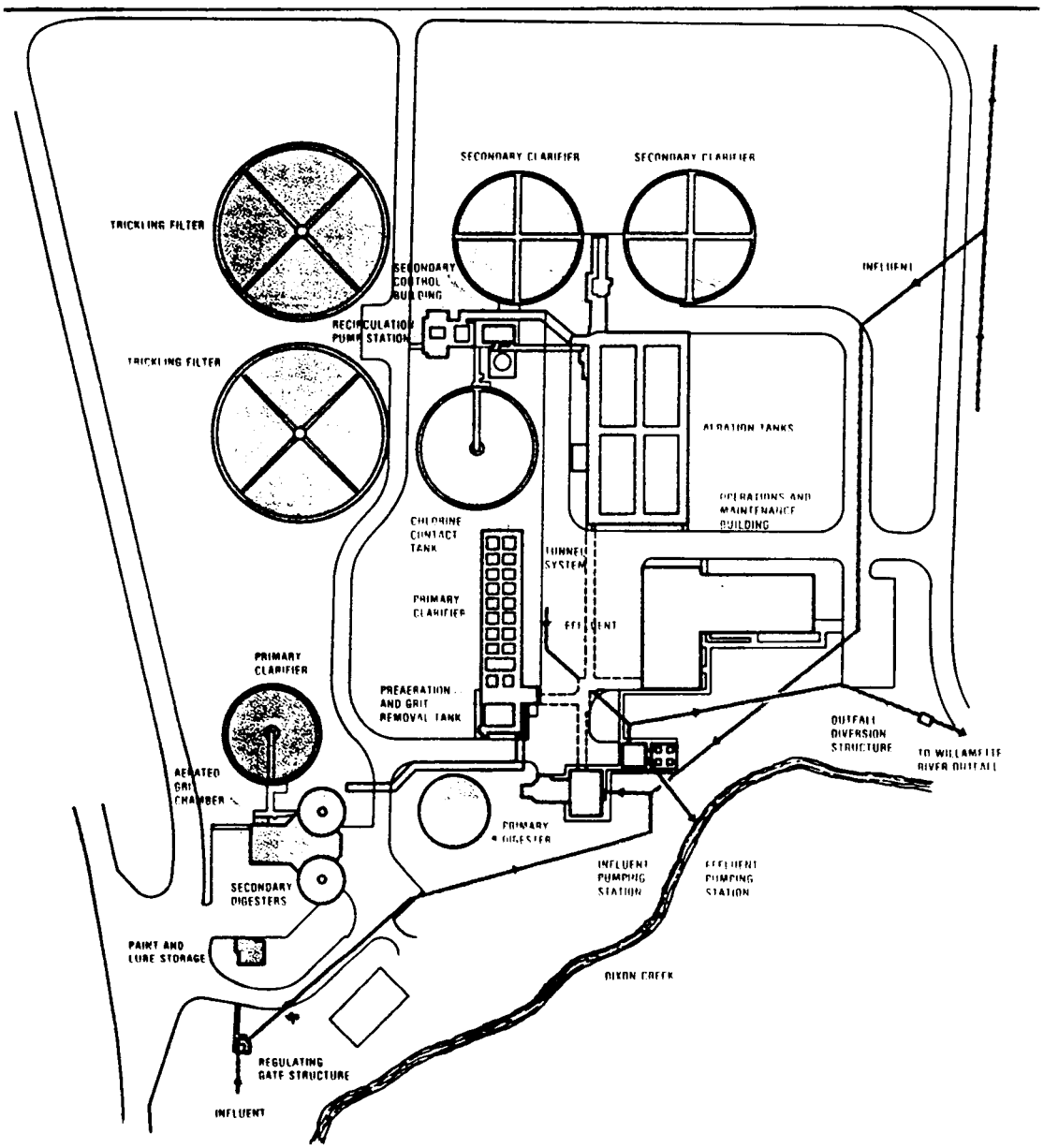

Figure 1. Corvallis Wastewater Reclamation Plant Systems and Layout (*City of Corvallis*, 1978)

sewer lines, or interceptors, that service more than 38,000 people. At the plant, the interceptors join to form a single 54-inch diameter pipe. Because no force other than gravity brings the raw sewage to the plant, the interceptor is located well below grade. The sewage flows to the influent pumping station where the pumps lift it 37 feet allowing it to flow throughout the treatment plant by gravity alone. The total pumping capacity is 28 million gallons per day, which is the factor limiting the total peak wet weather flow for the plant.

As the sewage leaves the pumping station, the flow is divided in two. Half goes to the older primary treatments systems and the other half to the newer primary treatment. The few differences between the treatment processes, other than age, involve different sequences and shapes. The old system draws the sewage through an aerated grit chamber, a comminutor, and a Parshall flume (a specially designed channel to measure flow) to a circular primary clarifier. The new system's sludge flows through a comminutor, a Parshall flume, and a preaeration-grit removal tank to a rectangular primary clarifier.

As mentioned in section 2.2.1, sand and other heavy inorganic particles settle in the aerated grit chamber and preaeration-grit removal tank. They are removed, washed, dewatered and deposited in a dumpster destined for a landfill. Once in the primary clarifiers, the sewage remains for one or two hours, allowing organic and inorganic suspended solids to settle (primary sludge) and floatable materials such as grease to rise. This primary sludge and scum are pumped to the anaerobic digester. Heretofore, all the processes have been a part of the primary treatment. Secondary treatment begins with the primary effluent flowing to the recirculation pump station and distributing it to a pair of

trickling filters. Built in 1965, these circular tanks are 160 feet in diameter filled with eight feet of rock. A slowly rotating distributor sprays the primary effluent over rocks which are covered with microorganisms that reduce the organic content by almost 50 percent.

After passing through the trickling filters, the filter effluent proceeds to the activated sludge units. Considered the second phase of the secondary treatment, these units consist of two rectangular aeration tanks and two circular secondary clarifier tanks. Naturally occurring microorganisms are also used in the activated sludge process to break down dissolved organic materials present in the wastewater. However, this process greatly differs from that of the trickling filters. A steady stream of activated sludge mixes with the filter effluent in the aeration tanks to form a mixed liquor. Compressed air agitates the mixed liquor and provides dissolved oxygen needed by the microorganisms to live. The bacteria in the activated sludge consume most of the remaining dissolved organic material during the two or three hour time span the wastewater passes through the aeration tanks.

Mixed liquor flows from the aeration tanks to the secondary clarifiers where the solids settle out as activated sludge. A portion of the activated sludge is reused in the aeration tanks to continue the cycle and is called return activated sludge. The excess is known as waste activated sludge which is returned to the primary clarifiers for removal to the digester. All sludge and scum in the system eventually reach the anaerobic digester.

The continuous anaerobic digestion process enables bacteria to break down the sludge and produces methane and carbon dioxide gases. Due to its status as a valuable

resource, the methane gas is used as fuel for heating the digester and other components at the CWRP. Retention of sludge in the anaerobic digester varies from about 21 to 30 days, depending on flows and time of year. The digested sludge is pumped to two 15 feet deep, 4.4 acre lagoons and allowed to settle. It remains there for up to two years until the sludge is removed and placed on agricultural fields.

The remaining step in the secondary phase of the treatment process involves the addition of chlorine solution to the clear effluent discharged from the secondary clarifiers. Upon chlorination, the effluent remains in a 110 foot diameter chlorine contact tank for a few hours to kill disease-causing organisms. The chlorine addition is automatically controlled to ensure the proper degree of treatment.

After flowing through the CWRP, the final effluent is of the very highest quality, exceeding Oregon Department of Environmental Quality (DEQ) and EPA standards. The plant protects the environment by removing more than 95 percent of the initial pollution. A portion of the plant effluent is reclaimed for utility service throughout the plant and irrigation of the plant grounds. The excess effluent is discharged to the Willamette River, and, in times of high water, a pumping station diverts the effluent to Dixon Creek, a Willamette tributary. (*City of Corvallis*, 1978)

2.4.3 Corvallis' Combined Sewer Overflow Status

Untreated sewage entering the Willamette River has always been a problem for the City of Corvallis, even with the existing CWRP. During dry weather, the plant adequately treats all sanitary sewage. However, in certain parts of town where systems for

wastewater and storm water are combined, wet weather flows exceed piping and plant capacity. The overflow containing untreated wastewater and storm water, also known as a combined sewer overflow (CSO), discharges into the Willamette River. In a typical year, 425 million gallons of the combined sewage and storm water are discharged to the river. (*Combined Sewer Overflow*, 1995a) Currently, the city is working to change this, prompted by federal and state regulations like the Clean Water Act. The DEQ has required Corvallis to control CSOs by December 2001. As a result, the city is proposing to expand treatment levels to include primary treatment of nearly all the wet-weather overflows. (*The City*, 1995)

On November 6, 1995, the city council approved a strategy for a CSO remediation entitled the First Street Relief Interceptor. The plan includes:

- New 42 and 36 to 72-inch diameter pipe along First Street from Western Boulevard to the wastewater reclamation plant
- 10 million gallon storage lagoon and pump station at the wastewater reclamation plant
- 35 million gallon per day CSO treatment facility at the wastewater reclamation plant
- Expansion by 10 million gallons per day of the existing wastewater reclamation plant

Including storage, pumps, conveyance, and treatment, the estimated cost for the First Street Relief Interceptor plan is $28,770,000. In order to meet the strict compliance schedule set forth in the agreement between the City of Corvallis and the DEQ,

preliminary engineering and other activities are already under way. (*Combined Sewer Overflow*, 1995)

2.5 BIOSOLIDS DISPOSAL

Although wastewater treatment plants vary widely in size, type of treatment, and system configuration, one aspect they all have in common is the generation of waste. The hardest part of implementing and maintaining a wastewater facility is the disposal of the removed material, particularly sludge. (Tchobanoglous, 1987)

2.5.1 Albany Biosolids Contributions

Of course large plants generate greater amounts of sludge than small plants. Large facilities also have sludge removal techniques that would not be appropriate for small wastewater treatment plants, and vice versa. Each plant is specifically designed to handle an estimated volume of sludge generated by the community. However, when a treatment plant becomes responsible for more sludge than anticipated, sludge removal becomes a high priority. This is true for the Corvallis Wastewater Reclamation Plant (CWRP).

In 1993, the City of Albany, Oregon, was no longer able to remove sludge generated by the city because their contract expired with a sludge removal service. Therefore Albany and Corvallis developed and signed a three year contract allowing the City of Albany to dispose of their sludge in the CWRP lagoons for a designated cost. Once every other day from October to June, the Albany Wastewater Treatment Plant fills

a tanker truck with sludge and transports it to the CWRP to be deposited in the storage lagoons. Unfortunately this procedure has put an undue burden on the storage lagoons, causing them to operate at 160% of their designed capacity. This in turn has removed the five to eight foot water cap which normally covers the biosolids that have settled. As a result, odor has increased and the nearby environment is exposed to nearly 6 million gallons of sludge due to the absence of the uncontaminated water barrier.

Another stipulation of the contract requires the City of Corvallis to assume all responsibility for the City of Albany's sludge. In order to prevent future final disposal implications, the incoming biosolids are often tested for nitrogen and heavy metals content. If the sludge does not meet the requirements, the CWRP has the right to refuse the shipment.

In December, 1996, the contract expires and once again Albany becomes responsible for the storage and ultimate disposal of their own sludge. It is uncertain at the time of this writing whether the contract will be renewed or allowed to end; no decisions have been made. (Clark, 1996)

2.5.2 Lagoon Storage and Disposal Sites

All the sludge the CWRP creates eventually passes through the anaerobic digester and is pumped to one of the two facultative storage lagoons. With a total surface area of 4.4 acres and a 15 feet depth, combined storage capacity of the lagoons exceeds 21 million gallons. This total was designed to include a seven to eight feet deep uncontaminated water cap, so the lagoons would ideally store a little more than ten million gallons of

biosolids. Under normal conditions, the lagoons would be able to store all CWRP sludge generated over five to six years without any removal. During the summer months, typically from June to October, the accumulated sludge is disposed of by application to local agricultural fields. No sense of urgency exists because the lagoons have plenty of capacity to store sludge generated throughout the winter. However this is not currently the case because of the Albany wastewater plant contribution. The lagoons exceed capacity, and it is necessary to empty one lagoon of approximately six million gallons each summer. Sludge removal has become a top priority. Even when using three tankers trucking 15-18 loads per day for 10 hours per day, seven days a week, the goal of emptying one lagoon during the summer remains unattainable.

2.5.2.1 Biosolids Removal Techniques

Sludge removal from the lagoons is not a complicated process. Seven days a week from June to October (actual dates are dependent upon amount of precipitation), local community college trainees drive a barge equipped with a vacuum pump systematically through the lagoons. The trainees slowly suck up the sludge which is deposited into a large white upright tank, resembling a silo. A gauge on the barge determines the solids content in the sludge, which should remain constant for proper field application. This is necessary for accurate record keeping of materials such as nitrogen and heavy metals applied to the fields (agronomic loading rates).

As sludge in the vertical tank increases, a boom is used to fill tanker trucks. The biosolids are then transported to the pre-determined fields and sprayed on top of the soil at

a constant rate. Practice and experience are necessary for the trainees and truck drivers in order to ensure an evenly distributed sludge volume on the fields with a constant amount of sludge content.

Incidentally, the City of Albany's digested sludge contribution during the summer application months is transported directly to Corvallis fields rather than pumping the sludge into the Corvallis lagoons. Even though it goes directly to the application sites, the CWRP remains responsible for the Albany sludge. Site location and amount of biosolids applied are still determined by and fall under CWRP contracts.

2.5.2.2 Characteristics of Application Sites

State approval is required before the application of sludge to fields, whether used for agricultural or soil restoration purposes. The CWRP must apply for a permit from the Oregon Department of Environmental Quality (DEQ) for each individual field. Field size, usage, soil and land characteristics must be provided on the permit application, employing Oregon State University guidelines to aid in determining these parameters. An approximation of heavy metals, nitrogen, and phosphorous loading rates are also included on the application. Upon DEQ approval of each individual field, the CWRP may legally apply digested sewage sludge generated from the plant. In order to ensure cost and efficiency, the CWRP applies biosolids to the same five or six fields each year, removing the hassle of new field analyses and permit applications.

As part of the DEQ guidelines, based on EPA 40 CFR 503 regulations, sludge applied to fields may not be closer than 200 feet from drinking water, 100 feet from open

water, and 50 feet from the site boundary. The EPA has extensively studied the effects of sewage sludge applied to agricultural land. "Development of Risk Assessment Methodology for Land Application and Distribution and Marketing of Municipal Sludge" is one of a series of reports that present sludge management practices. (EPA, 1989) The reports provide methods for evaluating health and environmental risks from toxic chemicals that may be present in sludge but do not address radioactive materials.

As per DEQ permit requirements, the timing and field usage of biosolids applications are regulated. Sludge must be applied after the field has already been harvested. Once the municipal sludge has been sprayed on the soil's surface, the land owner must plow and turn over the soil, lowering the possibility of runoff. Animals are not allowed to graze on the applied area for 30 days following the application and food crops cannot be harvested for 16-18 months.

Farmers, the CWRP, and the DEQ are all interested in knowing what materials are incorporated in the sludge. Farmers do not want their crops and soil contaminated or ruined and the DEQ wants to protect people and the environment from harmful chemicals. Therefore the CWRP monitors soil pH and employs an independent laboratory to analyze sludge for nitrogen, phosphorus, cadmium, nickel and other heavy metals and chemicals. A sample from the stabilized sludge in the lagoon is taken and analyzed four times throughout the sludge removal season (from June to October). The number of samples required directly relates to the total applied volume.

Using the results from the laboratory samples, the total amount of each chemical or metal applied to a particular field is recorded (in kg/ha) yearly. The Oregon DEQ has

determined limits for each constituent, and the amount of sludge applied to the fields is restricted by such loading rates. Once those limits have been reached, no matter how many years it takes, the site becomes disqualified for any further sludge application.

2.5.2.3 Corvallis Biosolids Application Sites

In addition to satisfying all Oregon DEQ requirements, an application site must meet certain CWRP criteria. These guidelines are not necessarily required; however they are used as eliminating factors. Initially, land owners volunteer for selection of their fields. Operators and wastewater workers perform a general visual inspection to determine if the field is adequate. They note field location, size and accessibility. Obviously a field must be near the plant and be able to accommodate an 80,000 lb. tanker truck by having structurally sound bridges, areas to turn the truck around, and driveable terrain. If the site meets these general criteria, the full Oregon DEQ permit application processes begins.

The farmer receives the sludge at no cost if the field is located within a ten mile radius of the treatment plant. If the field happens to be located farther than ten miles from the treatment plant, the land owner must pay additional transportation costs beyond the ten mile radius.

As previously stated, because the addition of new fields for application is a tedious, lengthy process, the same five to six sites have been repeatedly used for many years. Those that received biosolids in 1995, and for many past years, can be noted in Table 2. One area may include several separate fields, each with their own site number. For

instance, the airport location has nine different sites (numbered 37-45), each with a grass seed crop.

Owner	Total Acres	Crop	Location	Site No.
Anderson, Lloyd	25	Grass Hay	T11S, R5W, Section 14	10-14, 36
City of Corvallis	993	Grass Seed	T12S, R5W, Section 22, 27, 28, 33, 34 (Corvallis Municipal Airport)	37-45
Gray, Dennis	200	Grass Hay	T10S, R5W, Section 13, 24	60-69
Oregon State University	2000	Grass Hay, Oats, Barley	T10S, R5W, Section 23, 24, 25, 26 (OSU Beef Barn)	70-79
Sander, Gary	75	Christmas Trees	T13S, R5W, Section 2	30

Table 2. Corvallis Wastewater Reclamation Plant 1995 Biosolids Application Sites

3. METHODS, MATERIALS, AND EQUIPMENT FOR BIOSOLIDS MONITORING

The NRC has only inspected 15 of the approximate 1100 NRC licensees across the United States that may discharge radioactive material to sanitary wastewater reclamation plants, thereby determining if radionuclide concentration problems exist. (GAO, 1994) It is unknown how many of the 2000 agreement state licensees have or have not been inspected. (GAO, 1994) No matter the case, licensees may still legally discharge radioisotopes into the sanitary sewer so long as the average monthly concentration limits are met. With this fact, many studies have been performed analyzing the movement and reconcentration of various nuclides through wastewater treatment plants. It has also been shown that certain radionuclides tend to reconcentrate through sewage treatment plant processes, even though influent concentrations may be nondetectable. (Larsen, 1995; Prewitt and Glass, 1994; Stetar et al., 1993; Parrotta, 1991) Occasional monitoring of such possible reconcentration problems has become necessary.

Oregon State University currently has the means to evaluate possible radionuclide concentrations in Oregon generated biosolids, and the program developed here in this study is just the beginning of a future comprehensive analysis.

3.1 BIOSOLIDS MONITORING OBJECTIVES

Because of the possibility of detecting a wide variety of radioactive materials in sludge, a general monitoring procedure is used. Rather than trying to determine every

possible isotope present, test results show whether or not radiation exists at significant levels in biosolids. Therefore gross alpha, gross beta, and gamma spectroscopy are used. These tests indicate if further study and identification of individual isotopes are needed.

Determining the proper radiochemical techniques involved with gross alpha, gross beta and gamma analyses with sludge required consulting several commercial radiochemistry laboratories including Teledyne Isotopes, Midwest Laboratory in Northbrook, IL, CORE Laboratories in Casper, WY, and Analytical Resources Incorporated (ARI) located in Seattle, WA. Each laboratory uses the EPA's *Prescribed Procedures for Measurement of Radioactivity in Drinking Water*, (*Procedures*) Method 900.0 for gross alpha and beta and Method 901.1 for gamma emitting isotopes. Unfortunately, the *Procedures* are used for drinking water which contains much fewer solids. The actual testing process for radioactive materials in sewage sludge varies from lab to lab by slightly modifying or 'tweaking' the drinking water methods in *Procedures*, thereby making each lab's methods proprietary. However, radiochemist consultant Robert Gunther at ARI provided several useful hints with regard to modifying the EPA *Procedures* manual. In short, the procedures used in this study are based upon the *Procedures* manual with modifications to facilitate the increased solids content of biosolids.

The identification of gamma emitting radionuclides listed in *Procedures* are straightforward and no special techniques are required. However, determining accurate gross alpha and gross beta concentrations are more tedious, strictly from the fact that alpha and beta particles have short mean free path lengths compared to gamma rays.

Sludge solids greatly attenuate these particles and therefore must be accounted for with the use of transmission curves. A graph for alphas and a graph for betas account for the attenuation of the particles and ultimately vary their detectability as the amount of solids in the sample is varied. With these goals in mind, the creation of a biosolids monitoring methods is discussed.

3.2 LABORATORY EQUIPMENT AND SET-UP

Equipment and materials needed for the determination of radionuclide concentration were located within Oregon State University's Radiation Center building. In room B124, the actual radiochemical procedures were performed under a hood, while the proportional counter was located in room C120 and the Mettler balance in room C118. Proper precautions were taken during the transfer of radioactive materials from room to room and general lab safety procedures were followed.

3.2.1 Balance and Pipetter

For this study, the balance and P1000 pipetter used were not assumed to be properly calibrated, and therefore checks were performed to validate their accuracy. The balance was a Mettler, No. 281006, Type HGT, digCap 160 grams, OSU #148371. To test its calibration, a standard weight set, OSU #172983, provided the masses. The balance was initially zeroed each time there was a period of inactivity, and readings were always taken with the sliding glass doors fully closed. Upon zeroing the balance, 20, 200,

and 5000 mg masses individually verified the balance to be in exact working order to the ten thousands decimal place. However the balance reading had a tendency to drift a few tenths of a milligram shortly after a mass was in place. This was remedied by accepting the reading first measured as accurate and not waiting for the balance to begin to drift.

The pipetter was a Gilson Pipetman® P1000, No. C197020, with a label stating it had been calibrated November, 1995 by Rainin, Express Repair. Two separate methods were employed to verify the delivery amount at the 1.00 ml level. The first method involved four 2/3 drams capsules. The capsules were weighed using the calibrated Mettler balance; then a pipetted 1.00 ml of distilled water was added to each capsule and the lid closed. The capsules were then reweighed to determine the weight of the added volume of water. The second method used a small tray placed on the balance which was initially weighed. 1.00 ml volumes were added to the tray eight times, recording the total mass after each addition. At 70° F, the mass density of water according the CRC Handbook of Chemistry and Physics was compared to the measured values from both methods. (Lide, 1994) The average amount delivered from both types of calibrations at the 1.00 ml setting was 0.974 +/- 0.0053 ml. This value is used as the amount of solution added throughout this study.

3.2.2 Proportional Counter for Gross Alpha and Beta Measurement

The measurement of gross alpha and gross beta concentrations within CWRP sludge took place in room C120 using a windowless gas flow proportional counter

(GFPC), Unit III, manufactured by Nuclear Measurements Corporation, Model PC-4, Serial #7105 (115 V, 60 Hz). However the instrument had not been calibrated for over four years, and a full quality assurance was instituted using Radiation Center Health Physics Procedures (RCHPP) 9, entitled *Standard Quality Assurance Procedures for Laboratory Radiation Detectors* approved by Senior Health Physicist D. S. Pratt. It should also be noted that the permanent tray portion of the counting chamber was polished and cleaned using Nevr Dull® polishing agent. The instrument was chosen for its ability to measure low alpha activities, distinguish between alphas and beta particles, and accurately measure high count rates because of low resolving times. Full details involving the theory and general operation of a GFPC are not discussed here, assuming the reader already has this knowledge. However, Glenn Knoll's *Radiation Detection and Measurement* book may provide useful help. (Knoll, 1989)

Firstly, a voltage plateau curve was developed. P-10 gas, a mixture of 90% argon and 10% methane, was set at 5 psi and a standard Coleman® lantern mantle containing ^{232}Th was used to generate alpha and beta particles. The chamber was purged for an initial 100 second interval and subsequent 10 second purges between two minute counts were performed while varying the voltage from 750 volts to 2000 volts in 25 volt increments. The semi-log plot determining the alpha and beta plateau operating voltages of 1150 V and 1875 V, respectively, is shown in Figure 2.

Secondly, according to RCHPP 9, control limits were established for the use of the Unit III GFPC. A 60 Hz input check was performed at both the alpha and beta plateau voltages. Twenty, one minute counts were taken at each plateau to determine the average

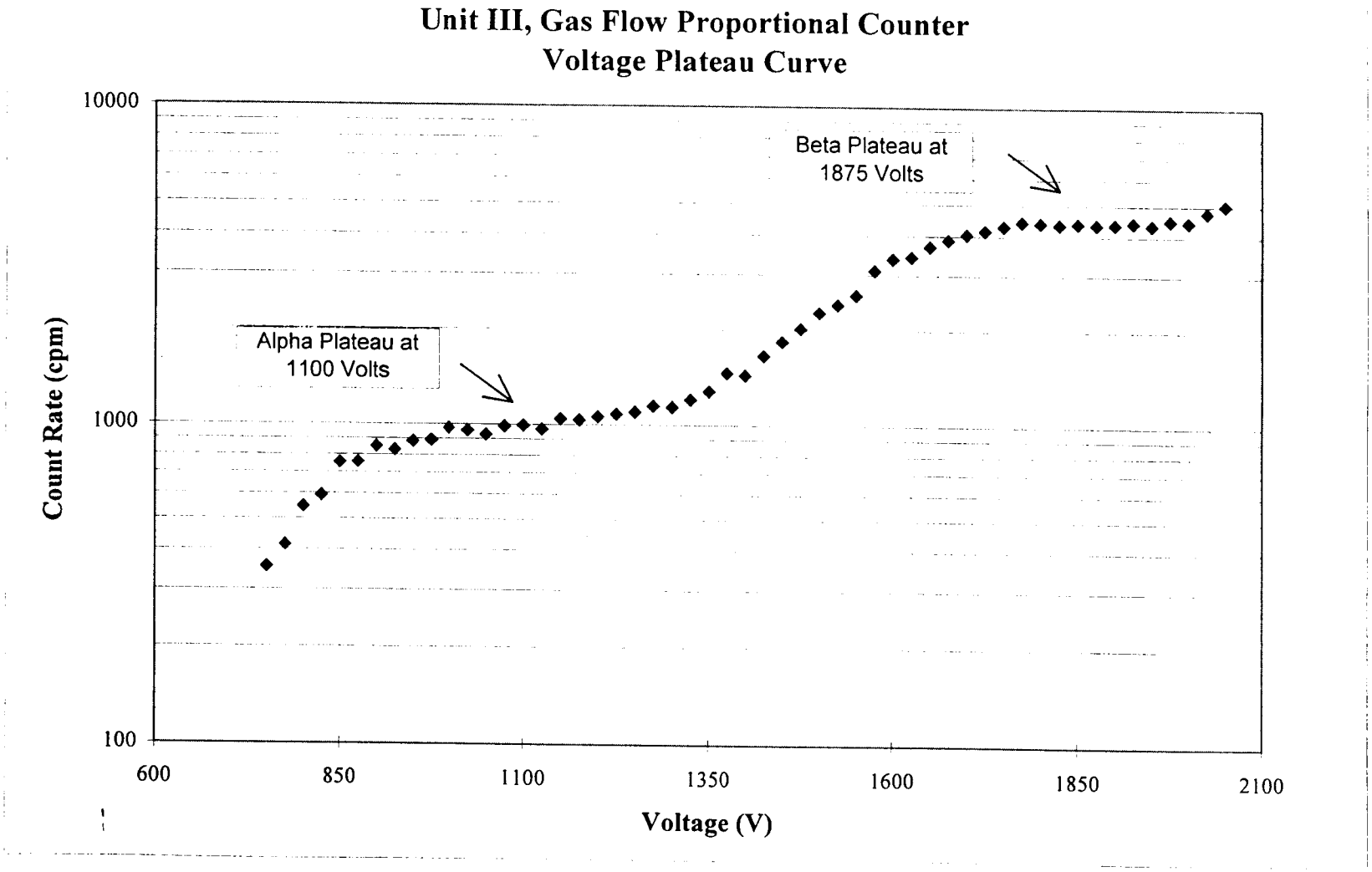

Figure 2. Unit III, Gas Flow Proportional Counter Plateau Curve

counts per minute. With a 95% confidence level, control limits were set from the entire population. Then twenty separate background counts were performed at 10 minutes each for both plateaus. In order to replicate experimental counting conditions, a clean planchet was placed in the counting tray and the chamber was opened and purged for 100 seconds each time. Again, control limits were set for future GFPC quality assurance using the standard deviation of the population at the 95% confidence level.

Thirdly, the GFPC Unit III resolving time was found using the two-source method. A ^{90}Sr source in two halves was counted using a 30 second purge for each count. Five separate one minute counts were recorded while analyzing each half of the source separately. Then another set of five one minute counts was performed with both halves together. Great care was taken in the source placement within the tray so as to maintain identical geometry for each count. The Unit III GFPC resolving time was found to be 0.104 microseconds.

3.2.3 Ge(Li) Detector

The ADCAM 3 Canberra lithium-drifted germanium gamma spectrometer module (detector #1643) located in B100 provided the means to perform a gamma spectroscopy on a biosolids sample using EG&G Ortec Maestro II computer software. Full details of the system configuration and theory are not discussed, although Knoll's *Radiation Detection and Measurement* may be referenced. (Knoll, 1989) Because a 500 ml polyethylene beaker would be used to count a sample of sludge, a standard reference Marinelli beaker OSU #16506 provided the necessary geometry similar to the test sample.

The NIST traceable source purchased from Amersham contained 11 different radionuclides with an overall uncertainty of 5%. However, since the standard reference date was on August 1, 1993, the shorter lived nuclides had decayed to background levels and were no longer detectable. However, the remaining nuclides such as ^{137}Cs and ^{60}Co still allowed the calibration of the system to be verified. An 80,000 second count using the Amersham Marinelli source generated a spectrum which was compared to the previously calibrated system. The photopeaks and associated energies were in very close agreement with the known values, and no changes had to be made in the calibration.

3.3 SLUDGE SAMPLING

For this study, a sludge sample was extracted directly from the sludge digester at the CWRP. Activated sludge enters the digester at the bottom of the one million gallon tank and the digested sludge exits near the top, where two one liter samples were taken. With the assistance of a CWRP operator, obtaining the samples involved changing the biosolids flow to a small valve which filled the sample containers. Because of stirring and mixing that takes place in the digester, the sample contents are considered representative of those in the tank.

Upon obtaining the samples, one liter of sludge for gross alpha and beta testing was preserved by bringing the pH level to near 2.0 with the use of concentrated nitric acid as recommended by *Procedures*, Method 900.0. A separate one liter sample for gamma spectroscopy was also preserved using concentrated hydrochloric acid and bringing the pH to approximately 2.0. The use of hydrochloric acid for preservation instead of nitric

acid for the gamma tested sample was recommended by Bob Gunther at ARI. He noted that by using nitric acid in the sample, any possible iodines present would no longer be detected. Previous testing was done to determine the amount of concentrated HNO_3 and HCl needed to bring a particular sludge sample to a 2.0 pH level. It was determined using new litmus paper on acidified sludge samples that each 100 ml of sludge requires approximately 0.87 ml of concentrated nitric acid and hydrochloric acid using Corvallis Wastewater Reclamation Plant digester sludge.

It was decided that short lived nuclides used at the local hospital, such as ^{131}I, would probably be detected more readily in relatively 'newer' digested sludge because the time taken for sewer line transport and treatment plant processes is much shorter than their half lives. This sample was not chosen as representative of the sludge that is applied to agricultural fields for three reasons. Firstly, the biosolids disposed on fields have been held in the storage lagoons for a few months at minimum, allowing the short lived radionuclides to decay. Secondly, with short lived isotopes included, a worst case scenario could be developed using all the isotopes originally contained in the recently digested sludge. Thirdly, obtaining a sample from the digester eliminated the need to row a small boat to the middle of the storage lagoon and scoop a settled sample.

Biosolids may be sampled throughout the year at most wastewater treatment plants. However, sampling the sludge as it is being poured into transport vehicles is quite simple for those reclamation plants which dispose of their generated biosolids off site. A direct determination can be made as to the amounts of radioactive materials actually put in the soil or landfill by examining sludge that leaves the plant. This type of sampling method

was not performed for the study because biosolids were not being transported at the time and because of the reasons previously mentioned.

One major assumption which provides statistically viable results is that the contents in the sample obtained be representative of the entire sludge volume. However this is not the actual case, but the assumption provides a close approximation. This valid assumption relies upon the normal continuous mixing and constant solids mass applied to agricultural fields.

3.4 GROSS ALPHA AND BETA DETERMINATION

Techniques involving the testing of preserved biosolids for gross alpha and beta were relatively simple. Approximately one liter of sample was collected at the CWRP and preserved by slowly adding 8.7 ml of concentrated nitric acid. Before removing smaller aliquots, the preserved sample taken from the treatment plant was shaken vigorously to create a uniform mixture of solids and liquids. Typically, a few milliliters were extracted and placed in a small closeable container such as a liquid scintillation vial. One milliliter of concentrated nitric acid was added to the vial in order to dissolve the solids content and subsequently promote an evenly distributed sample on a counting planchet. After the acid dissolved the solids, approximately one milliliter was removed and placed on a tared ringed stainless steel counting planchet and fully distributed throughout the surface. The planchet was then fully dried under an Infra-Rediator heat lamp for at least six hours. After weighing the planchet to determine the amount of residual solids, the sample was then ready to be counted using a proportional counter.

3.5 TRANSMISSION CURVE PREPARATION

Once the test biosolids were obtained, separate transmission curves for alpha and beta were generated by plotting transmission with units of counts per minute divided by disintegrations per minute (cpm/dpm) versus residual sample solids (in milligrams). In other words, radioactive material was added to samples with varying solids content, each sample counted, and the results plotted. The preparation and methods used to produce these separate graphs are as follows.

3.5.1 General Spiked Sample Preparation

As previously stated, the goal for this section is to create transmission curves using samples containing different amounts of solids with known alpha and beta activities. Accomplishing this task involved several steps. Ten small vials (liquid scintillation counter vials) were used to contain a mixture of several constituents and allow adequate mixing. Into 9 of the 10 vials, approximately 2 ml of sludge was added using a disposable plastic pipetter. The concentration of sludge solids within the vials was then varied by either adding different amounts of distilled water or concentrated, dewatered sludge. Concentrated sludge was used to add solids rather than a dissolved salt mixture as recommended by the EPA *Procedures* because inconsistencies have been found with different salt mixtures as was noted by a memo from the US EPA National Exposure Research Laboratory, Office of Research and Development. The method used to

concentrate the sludge involved centrifuging a normal sample and removing the excess water. The mass density of the concentrated sludge was measured.

A known amount of spike was then added to all 10 vials. In addition, a milliliter of concentrated nitric acid was place in each vial so that the solids component would dissolve and be distributed to a planchet more easily. The vials were lightly shaken and then left alone for the acid to fully dissolve the solids.

Twenty stainless steel 10 cm diameter ringed planchets were obtained to distribute the spiked, dissolved sludge. Using a rubber policeman, 1 ml was extracted twice from each vial and placed onto two separate planchets. Evenly distributing the liquid on the planchets was cumbersome and sometimes required a pipetter tip to break the surface tension and promote spreading. Any liquids left on the pipetter tip were wiped back onto the planchet. With the liquid fully distributed across the planchet area, the samples were dried under a hot lamp for at least six hours. (This length of time was adequate for complete dryness.) The above process was used for each isotope.

Therefore, the final result included 20 dried ^{241}Am spiked planchets and 20 dried ^{90}Sr spiked planchets, each with differing amounts of solids. Actual amounts added to the vials can be seen in Table 3. for the ^{241}Am vials and Table 4. for the ^{90}Sr vials.

3.5.2 Varying Transmission Curve Residual Solids

Both the alpha transmission curve and beta transmission curve necessitate a range of solids remaining on the planchet once it is dried. In order to properly vary the amount of solids present in sludge and subsequently on planchet, a starting point is needed with a

Vial Label	Concentrated Sludge (ml)	σ	Typical Sludge(ml)	σ	^{241}Am Spike (ml)	σ	Concentrated Nitric Acid (ml)	σ	Distilled Water (ml)	σ	Total Volume (ml)	σ
1-1,2	0	0	0	0	0.974	0.0053	0.974	0.0053	0.974	0.0053	2.921	0.009
1-3,4	0	0	2	0.1	0.974	0.0053	0.974	0.0053	3.894	0.0213	7.841	0.103
1-5,6	0	0	2	0.1	0.974	0.0053	0.974	0.0053	2.921	0.0160	6.868	0.102
1-7,8	0	0	2	0.1	0.974	0.0053	0.974	0.0053	0.973	0.0053	4.921	0.100
1-9,10	0.12	0.006	2	0.1	0.974	0.0053	0.974	0.0053	0	0	4.067	0.101
1-11,12	0.30	0.015	2	0.1	0.974	0.0053	0.974	0.0053	0	0	4.247	0.101
1-13,14	0.48	0.024	2	0.1	0.974	0.0053	0.974	0.0053	0	0	4.427	0.103
1-15,16	0.66	0.033	2	0.1	0.974	0.0053	0.974	0.0053	0	0	4.607	0.106
1-17,18	0.84	0.042	2	0.1	0.974	0.0053	0.974	0.0053	0	0	4.787	0.109
1-19,20	1.02	0.051	2	0.1	0.974	0.0053	0.974	0.0053	0	0	4.967	0.113

Table 3. Vial Contents for ^{241}Am

Vial Label	Concentrated Sludge (ml)	σ	Typical Sludge(ml)	σ	^{241}Am Spike (ml)	σ	Concentrated Nitric Acid (ml)	σ	Distilled Water (ml)	σ	Total Volume (ml)	σ
2-1,2	0	0	0	0	0.974	0.0053	0.974	0.0053	0.974	0.0053	2.921	0.009
2-3,4	0	0	2	0.1	0.974	0.0053	0.974	0.0053	3.894	0.0213	7.841	0.103
2-5,6	0	0	2	0.1	0.974	0.0053	0.974	0.0053	2.921	0.0160	6.868	0.102
2-7,8	0	0	2	0.1	0.974	0.0053	0.974	0.0053	0.973	0.0053	4.921	0.100
2-9,10	0.14	0.007	2	0.1	0.974	0.0053	0.974	0.0053	0	0	4.087	0.101
2-11,12	0.35	0.018	2	0.1	0.974	0.0053	0.974	0.0053	0	0	4.297	0.102
2-13,14	0.56	0.028	2	0.1	0.974	0.0053	0.974	0.0053	0	0	4.507	0.104
2-15,16	0.77	0.039	2	0.1	0.974	0.0053	0.974	0.0053	0	0	4.717	0.107
2-17,18	0.98	0.049	2	0.1	0.974	0.0053	0.974	0.0053	0	0	4.927	0.112
2-19,20	1.19	0.060	2	0.1	0.974	0.0053	0.974	0.0053	0	0	5.137	0.117

Table 4. Vial Contents for ^{90}Sr

normal unspiked sample. To determine the amount of solids present in the normal sludge, preserved with nitric acid, 1 ml amounts were removed and placed on 10 tared planchets. The wet samples were again weighed using a calibrated Mettler balance No. 281006, type HGT, OSU #148371 and then dried using a Fisher Infra-Rediator, Model 11-504-5V4 for approximately 6 hours. With all of the liquid content evaporated, the samples were again weighed. By a ratio of masses the sludge samples were calculated to be approximately 3% to 4% solids, with 1 ml of sludge containing on average 13 mg of solids. The solids content for the spiked samples used in the generation of the transmission curve was then varied around the 13 mg midpoint.

3.5.3 Spiking the Biosolids

Sludge with varying solids is spiked to create the attenuation characteristics needed for correct transmission curves. Even though any type of radioactive material could have been used to generate attenuation curves, ^{241}Am was selected as the best isotope for the alpha curve and ^{90}Sr the best for the beta curve for several reasons.
Even though the ^{241}Am 5.49 MeV alpha particle energy is higher than those emitted by naturally occurring uranium and ^{226}Ra, it is similar to the alpha energies emitted by naturally occurring ^{228}Th and ^{224}Ra and therefore produces a similar attenuation effect. ^{241}Am has also been previously detected in sewage sludge because of its widespread use in smoke detectors causing necessary remediation incidents. In the past, ^{90}Sr has also been found in sewage sludge. It is readily available and the energetic 2.28 MeV beta from the

^{90}Y daughter provides an adequate high energy beta to the 0.546 MeV ^{90}Sr beta. Finally, both isotopes were recommended for use by the EPA *Procedures* manual.

Standard solutions were purchased from Isotope Product Laboratories in Burbank, CA. Each solution arrived in a flame sealed ampoule containing 5 ml of 0.1 M HCl acid. Both ^{241}Am and ^{90}Sr standards are National Institute of Standards and Technology traceable, based on the blind assay of Standard Reference Materials as in NRC Regulatory Guide 4.15. Table 5. contains a summary of each standard isotope.

3.5.4 Dilution of Standards

The approximate 100 μCi standards of ^{241}Am and ^{90}Sr ordered from Isotope Products Labs came in a 5 ml flame sealed ampoule. Manipulating and working with the radionuclides required that each ampoule be slowly scored with a file until the tip broke off. The upper part above the neck of each glass ampoule was marked with a gold line, however no instructions came with proper opening procedures or a reason for the gold line. Assuming the line indicated the optimum scoring location on the ampoule, the ^{241}Am was broken open and the contents transferred to a flask. Rigorous shaking of the ampoule was needed to break the surface tension of the liquid. Because of these problems, the ^{90}Sr ampoule was scored and broken open at the more convenient location at the neck. Removing the contents was even more difficult due to the small diameter of the opening.

Both ampoules were rinsed using a 0.1 M HCl solution at least four times with the rinse solution added to the flask. Each flask was filled to the 100 ml mark +/- 0.06 ml at

	^{241}Am	^{90}Sr
Half Life	432.22 +/- 0.66 years	28.5 +/- 0.2 years
Isotope Product Laboratories Catalog No.	7241-100U	7090-100U
Reference Date	1 May 1996	1 May 1996
Contained Radioactivity	96.40 μCi (3567 kBq)	100.6 μCi (3723 kBq)
Mass of Solution	5.05779 g in 5 ml	4.99964 g in 5 ml
Chemical Form	$AmCl_3$	$SrCl_2$
Carrier Content	10 μg Eu/ml	10 μg Sr +50 μg Y/ml
Density	1.0171 g/ml @ 20°C	0.9996 g/ml @ 20°C
Radionuclide Concentration	19.06 μCi/g	20.12 μCi/g
Measurement Uncertainty		
Instrument Calibration	+/- 3.0%	+/- 1.5%
Assay	+/- 1.3%	+/- 1.6%
Weighing	+/- 0.0%	+/- 0.0%
Total at 99% Confidence Level	+/- 3.3%	+/- 2.2%

Table 5. Characteristics of Isotope Standards

20° C with additional 0.1 M HCl acid to prevent the isotopes from plating out or precipitating onto the inner walls of the Pyrex® flask.

Counts were still measured on the empty ampoules using an Eberline Instrument Corp. Geiger-Mueller detector, Model RM-3A, EBGM588, S/N 588 for the ^{90}Sr standard so a check giving a rough estimate of the remaining activity was performed. 12,000 cpm at a 22% efficiency showed 24.6 pCi (909.1 Bq) remaining or 0.024% of the original activity. The same analysis was performed with the ^{241}Am ampoule using a Ludlum Measurements Incorporated alpha detector, Model 28, S/N 5242. The remaining activity was 1.3 pCi (48.6 Bq) or 0.0014% of the total activity.

With the above procedures and considerations regarding the dilution of the standards, the final concentrations of standard solutions used for the study were

- ^{241}Am --- 0.964 +/- 0.032 μCi/ml
- ^{90}Sr --- 1.006 +/- 0.022 μCi/ml

These values include the errors associated with the standard activity from Isotope Products Labs and the flask solution volume.

To spike the biosolids, 1 ml of standard solution at the above concentration was added to a vial to make alpha and beta samples, respectively. Such high activities were used to overcome any possible activity that may have been already present in the unspiked biosolids. If very small activities were used as spikes, outcomes would be biased from the presence of existing radionuclides in sludge. Addressing this bias meant performing a standard additions method to find the activity levels without any spike. This problem was avoided by using high activities that would not be influenced by very small amounts of

existing activity. No matter the level of activity used as a spike, the alpha and beta attenuation characteristics remain the same; therefore the shape of the transmission curve developed would be the same.

3.6 GAMMA COMPONENT EVALUATION

Testing for particular gamma emitting radionuclides in the preserved unspiked biosolids was accomplished using gamma spectroscopy. The collected sample was immediately preserved by adding concentrated HCl acid solution to obtain a sample pH of 2.0. Preservation with nitric acid would have driven off any detectable iodines within the sample, therefore hydrochloric acid preserved any present isotopes including iodine. A 400 ml sample placed in a Marinelli beaker contained approximately 3.5% solids and was rigorously shaken in order to adequately mix the solids component into a uniform density. Unfortunately, during the 80,000 second count the solids settle and therefore change the mixture to a nonuniform distribution. This action should only minimally affect the detector gamma efficiency because of the penetrating characteristics of gamma rays in conjunction with the limited detector capability of distinguishing the low energy gamma normally attenuated. In general, the gamma spectrometer is applicable for analyzing gamma energies from 60 to 2000 keV.

Once the gamma spectrum of the unspiked biosolids was saved, an 80,000 second background count was performed and saved. A discussion of the minimum detectable activities and results for the gamma spectroscopy are later presented.

4. RESULTS

4.1 PROPORTIONAL COUNTER ANOMALIES

Generating the transmission curves required counting 20 ^{241}Am planchets several times each at the alpha and beta plateau voltages and counting 20 ^{90}Sr planchets several times each at the beta plateau voltage. With such numerous counts, filter paper was placed in the bottom tray of the Unit III proportional counter to prevent possible contamination. Each time the tray was opened and closed to change a sample the oxygen content in the counting volume was replaced with P-10 gas with a 100 second purge. Ten second purges were used after each two minute count when the tray was not opened.

These counting techniques were proven problematic when the data were analyzed. The irregular and peculiar results led to an investigation of possible factors. The Unit III GFPC cover was removed, exposing a pair of unattached springs used to provide a tight chamber seal. Needless to say, the springs were reattached. A rubber seal was also examined, lubricated and determined adequate. Tests then were implemented using single standard disk and planchet samples which were counted repeatedly on Unit III, Model PC-4 (1975), Unit IV, Model DC-4 (1975), and Unit V, Model PC-5 (1978) proportional counters. Count rates widely varied for a single sample without any apparent physical changes. Longer count times were eliminated due to the possibility of introducing unseen variations in count rate. Even after numerous tests accounting for each variable, the resulting count rates remained obscure and abnormal. However, the several following conclusions were still drawn.

- The proportional counter's power supply and high voltage should always remain turned on to prevent internal circuitry changes, contrary to the manual's instructions.
- At minimum, a 30 second purge introducing fresh gas should be used between each count where the tray door was unopened.
- Filter paper under the sample in the tray causes erroneous results and should not be used. Every disk source or sample should remain in full contact with the tray to prevent an influencing static charge.

According to the EPA *Procedures* manual, samples have a tendency to become hygroscopic as a result of the nitric acid dissolution of some types of organics. This introduction of water into the sample, or the possibility of the sample not being fully dried, would cause erratic counting and abnormal attenuation curves. However, these scenarios were checked repeatedly. Dissolved sample weights on the planchets remained constant over several weeks, and could not have contributed to the counting problems.

The planchets were recounted on GFPC Unit III drawing on the conclusions previously mentioned. Thirty second purges between counts were used, the counter power and high voltage were continually on, filter paper under the sample was eliminated, and each count time was held at one minute. The following results utilized these changes.

4.2 TRANSMISSION CURVES

Transmission curves for ^{241}Am and ^{90}Sr account for attenuation of the respective alpha and beta particles by the dried solids on the planchet. However, two planchets for

each radionuclide solely contained distilled water, concentrated nitric acid and a spike. Sludge was purposely withheld to allow these planchets to act as standards allowing the calculation of individual proportional counter efficiencies. The remaining spiked solids planchets develop the attenuation curve characteristics. Therefore, the particular transmission curves shown are also efficiency curves for the Unit III GFPC alone. If a different detector (with a its own unique absolute efficiency) were to be used for the analysis of unspiked sludge samples, the transmission curves would remain applicable. The planchets without any biosolids and a known amount of activity would be counted and the proportional counter efficiency calculated. The transmission curve could be adjusted or 'slid' to the new efficiency level because the general shape of the attenuation curve remains unchanged.

4.2.1 Alpha and Beta Curves

Determination of each individual point plotted on the transmission curves originated from weight measurements and detector counts. The weight measurements were straightforward, finding the residual solids in milligrams. Alternatively, a few detector counts grossly varied even after the changes in counting procedures. However, every one minute count result was included in the analysis. For those planchets with similar consecutive count rates, only a few counts were taken. Other planchets were recounted until the count rate leveled off at some particular value. One planchet was recounted 13 times with all results recorded. Every count for each planchet was used to determine its overall average value and standard deviation. This method gave quite a

spectrum of standard deviations for each sample, but still honestly included every measured value. Count rates were then divided by each planchet's computed activity to find an efficiency. As previously stated, each pair of planchets originated from a particular vial with a known activity and solids constituent. This allowed the results from each pair of planchets to be averaged, changing the number of data points from 20 to 10. Of course the standard deviation from each planchet was propagated.

Figure 3. displays the curve used to account for the attenuation of alphas in biosolids and Figure 4. attenuation for betas. A biosolids sample would be prepared using the procedures previously mentioned and subsequently counted on a detector whose efficiency and background are known at the appropriate voltage plateau. The desired minimum level of activity depends on the length of time counted, which is discussed in section 4.3.1. Once the sample is counted, the counter efficiency and attenuation effects are resolved by using the transmission curve. By knowing the mass of planchet residue solids, an absolute efficiency is easily determined for that particular sample. The curves are only valid for samples with less than 25 milligrams residue solids per milliliter. Samples with a high solids content should be diluted with distilled water to obtain the necessary mass.

The ^{90}Sr curve contains six, rather than ten, data points because anomalous results, which could not be accounted for, were found for those planchets with residue masses greater than 25 mg.

Once again it should be noted that the presented curves ideally apply only to ^{241}Am and ^{90}Sr. These isotope's emitted particles have unique energies which determine their

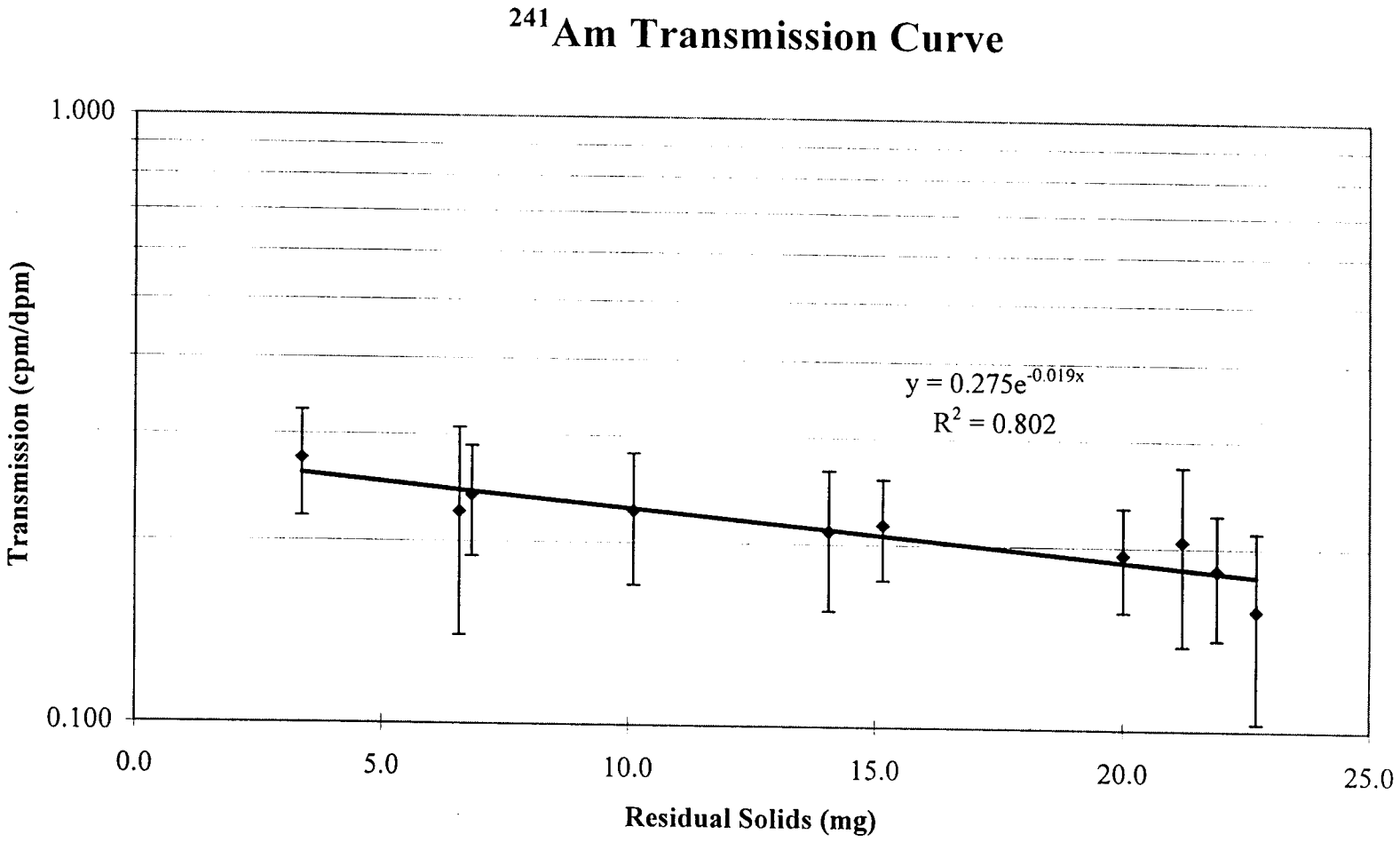

Figure 3. ^{241}Am Transmission Curve

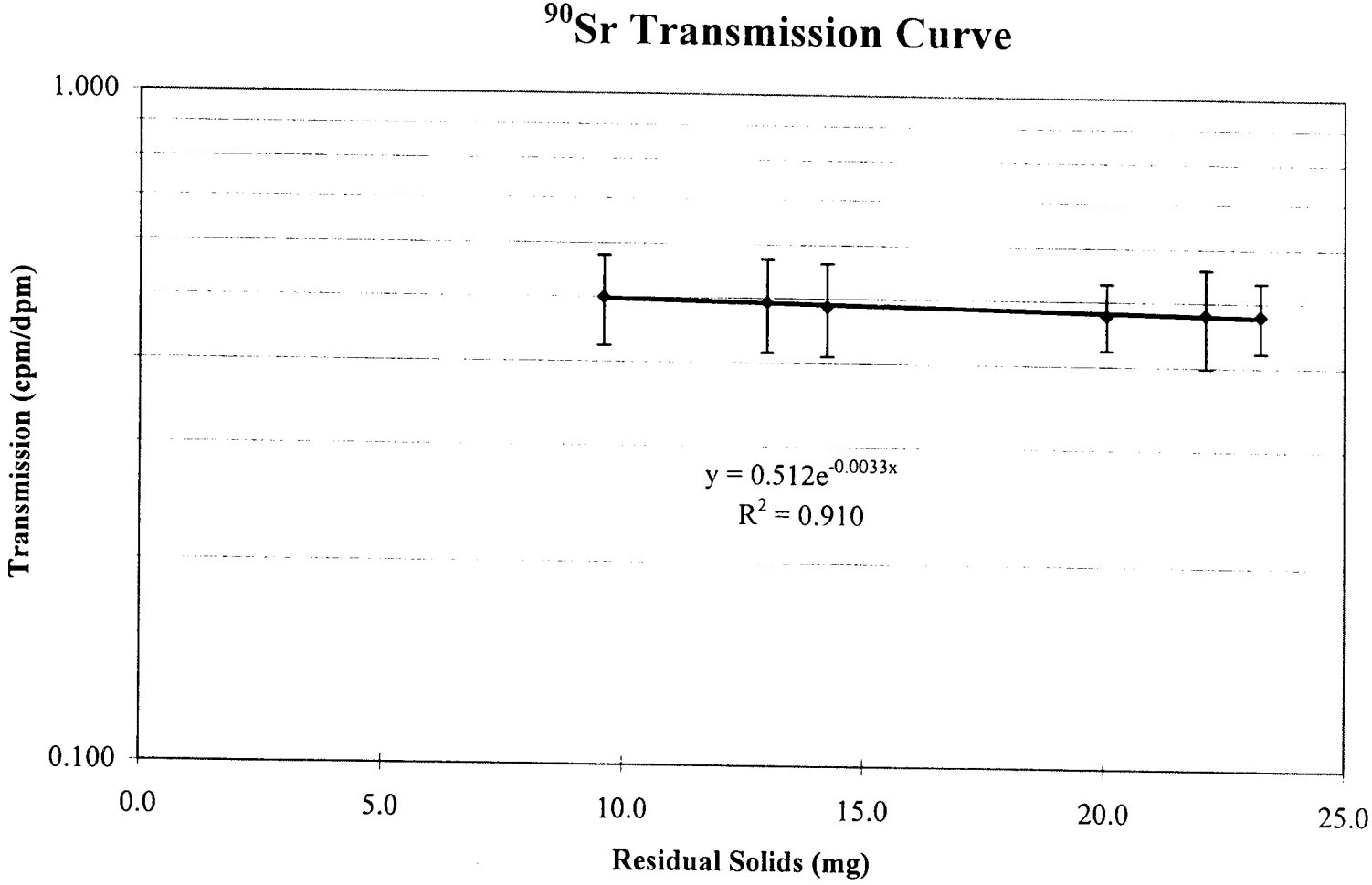

Figure 4. ^{90}Sr Transmission Curve

own attenuation characteristics. Nevertheless, the transmission curves may be used as general nuclide detection tools, determining if an in depth evaluation or isotopic analysis is necessary.

4.2.2 Sample Error

Generating the transmission curves to account for alpha and beta attenuation involved many steps, each with an associated error. From the original standard solution error, to the error associated with liquid measurements, to detector counting errors, each must be accounted for and propagated through every calculation. The error bars attached to the curve reflect the total error throughout the study at a 95% confidence level. The largest error for data point efficiency on the ^{241}Am curve was +/- 8.4%, and for the ^{90}Sr curve +/- 8.0% with 95% confidence. The main contributor to the total error for each data point originated from the procedure which removed the spiked solids from the vial and deposited it on the planchet. A more precise measurement of the total liquid volume could not be achieved because the pipetter was unable to extract a solution with such a large solids content. Instead a 10 ml disposable transfer pipette and rubber policeman withdrew about 1 ml +/- 0.05 ml of vial mixture.

The standard error of the absolute efficiency was calculated for each curve and found to be 1.4% for the alpha curve and 0.3% for the beta curve. These values incorporate errors associated with the fitted curve coefficients and subsequently give the total error associated with any future results read from the curves pertaining to the absolute efficiency.

4.3 SAMPLE COUNTING STATISTICS

If a sample contains activity which nears background levels, it becomes difficult to determine if the counts originated from background variations or the activity present in the sample. The distribution of counts from background and actual activity overlap and can produce either a false negative (type I) error, saying no activity is present when in reality there is, or a false positive (type II) error, deciding activity is present when actually it is not. By using a 95% confidence level ($\alpha = 0.05$), a lower detection level is set as a function of background standard deviation from a specific counting time.

4.3.1 ^{241}Am and ^{90}Sr Minimum Detectable Activities

Assuming a Poisson distribution for counting radioactivity, the distribution is asymmetrical at low mean values, requiring a slight increase in the correction factor. Including detector absolute efficiency and count time, the formula used for minimum detectable activity (MDA) suggested by NUREG 1156 is

$$MDA = \frac{4.65\sigma_b + 3.0}{\varepsilon_a t}$$

where

σ_b = standard deviation of the detector's background count

ε_a = absolute efficiency in cpm/dpm

t = background counting time

By using this formula and the equations for the lines fitted on the transmission curves, a gross alpha and beta MDA can be determined for any biosolids sample with mass from 0 - 25 mg.

From the ^{241}Am curve,

$$MDA_{\alpha} = \frac{4.65\sigma_b + 3.0}{t \cdot 0.275e^{-0.019(\text{residual solids (mg)})}}$$

From the ^{90}Sr curve,

$$MDA_{\beta} = \frac{4.65\sigma_b + 3.0}{t \cdot 0.512e^{-0.0033(\text{residual solids (mg)})}}$$

These formulas may be used for any sample counted on the Unit III GFPC at the respective voltage plateaus. If a different detector were used, the spiked planchets without biosolids would be first counted to determine the absolute efficiency, the curve adjusted, and the new MDA equation used.

4.3.2 Counting Time Optimization

A counting procedure to determine a sample net count rate is normally carried out by counting it for a specific period of time and then subtracting the background component. The net count rate found has a particular standard deviation which is determined through error propagation. If a total time to determine background count rate and net sample count rate is determined, the standard deviation can be minimized by choosing the best fraction of total time allocated for sample and background count times.

Through squaring and differentiating the equation for standard deviation, the following formulas can be used for the optimum division of time. (Knoll, 1989)

$$\frac{T_{S+B}}{T_B} = \{(S+B)/B\}^2$$

and

$$T = T_{S+B} + T_B$$

where

T = fixed total time

T_{S+B} = source with background count time

T_B = background count time

S = net count rate

B = background count rate

4.3.3 Error Propagation

Most steps for this study involved some type of error. Therefore, error propagation was used to find the correct value for the standard deviations. The two main formulas used to continually include and update the current standard deviations were:

for addition and subtraction

$$\sigma^2_t = (\sigma^2_x + \sigma^2_y + \ldots \sigma^2_n)$$

and for multiplication and division

$$\frac{(\sigma_t)^2}{(t)^2} = \frac{(\sigma_x)^2}{(x)^2} + \frac{(\sigma_y)^2}{(y)^2}$$

As each step was calculated, the proper error propagation formula was used. Of course, sigma represents the standard deviation of the estimated value.

4.4 LITERATURE COMPARISON WITH ^{241}AM

In November 1995, the EPA released a note mentioning problems with attenuation curve results generated from water samples. A bias had been discovered in the gross alpha methods used since 1980. For water studies, the amount of residual solids were varied by the addition of dissolved salt solutions to the samples. Using the same isotope but different salt solutions, variations of 50 to 70 percent were found in the 30 to 40 mg range. Therefore discovering the material used to vary sample solids has a direct impact on transmission curve shape.

The results obtained from this study's alpha transmission curve were compared to those from the EPA National Exposure Research Laboratory in Las Vegas, NV. (EPA, 1995) The EPA used a sulfate salt solution to change the amount of residual solids while this study used concentrated biosolids. Consequently, the curves were expected to be somewhat different because of the newly found EPA study bias. This comparison is made for general agreement purposes only. It is also assumed the detector efficiencies are different, as shown in Figure 5.

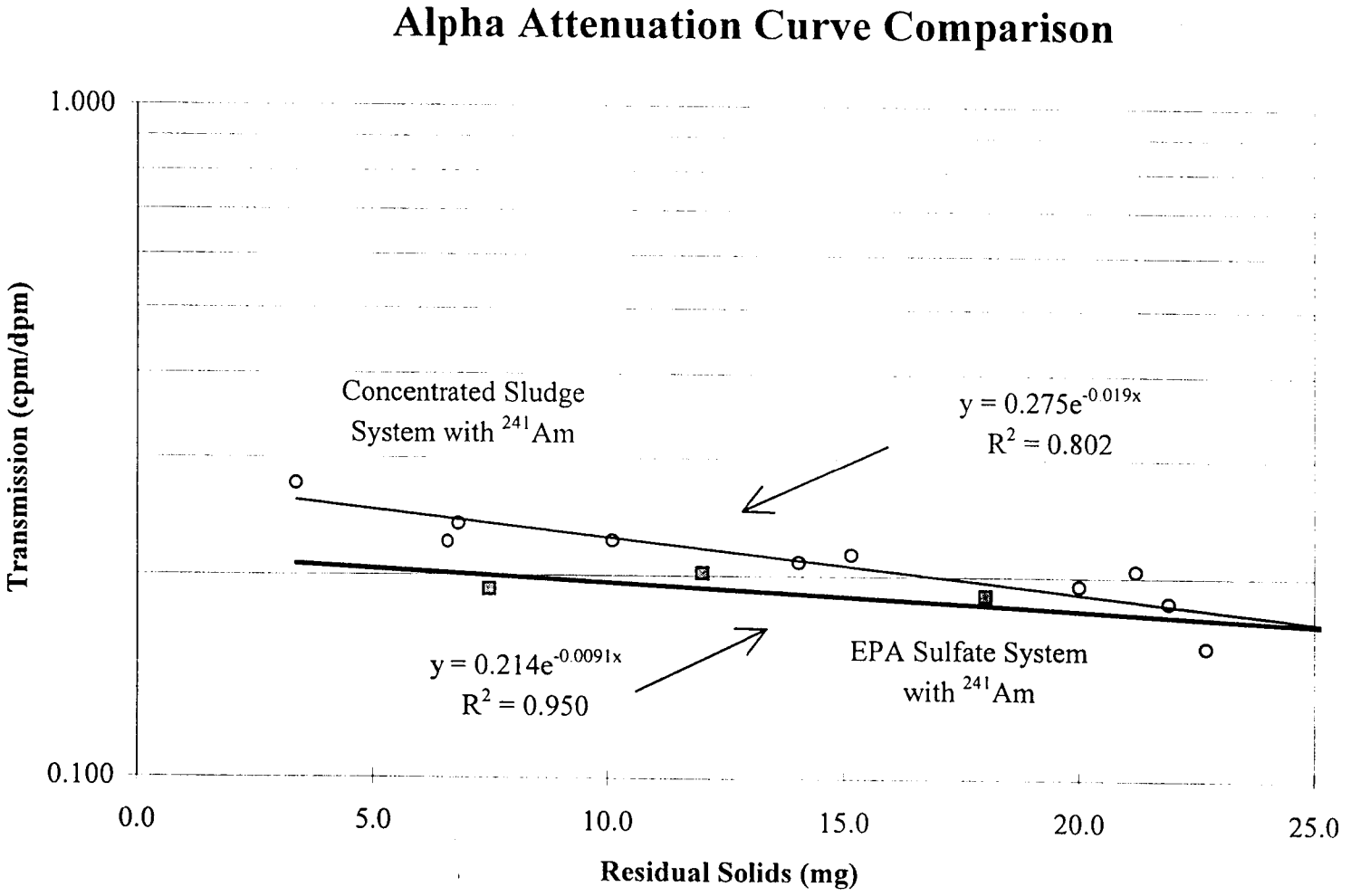

Figure 5. ^{241}Am Transmission Curve Comparison

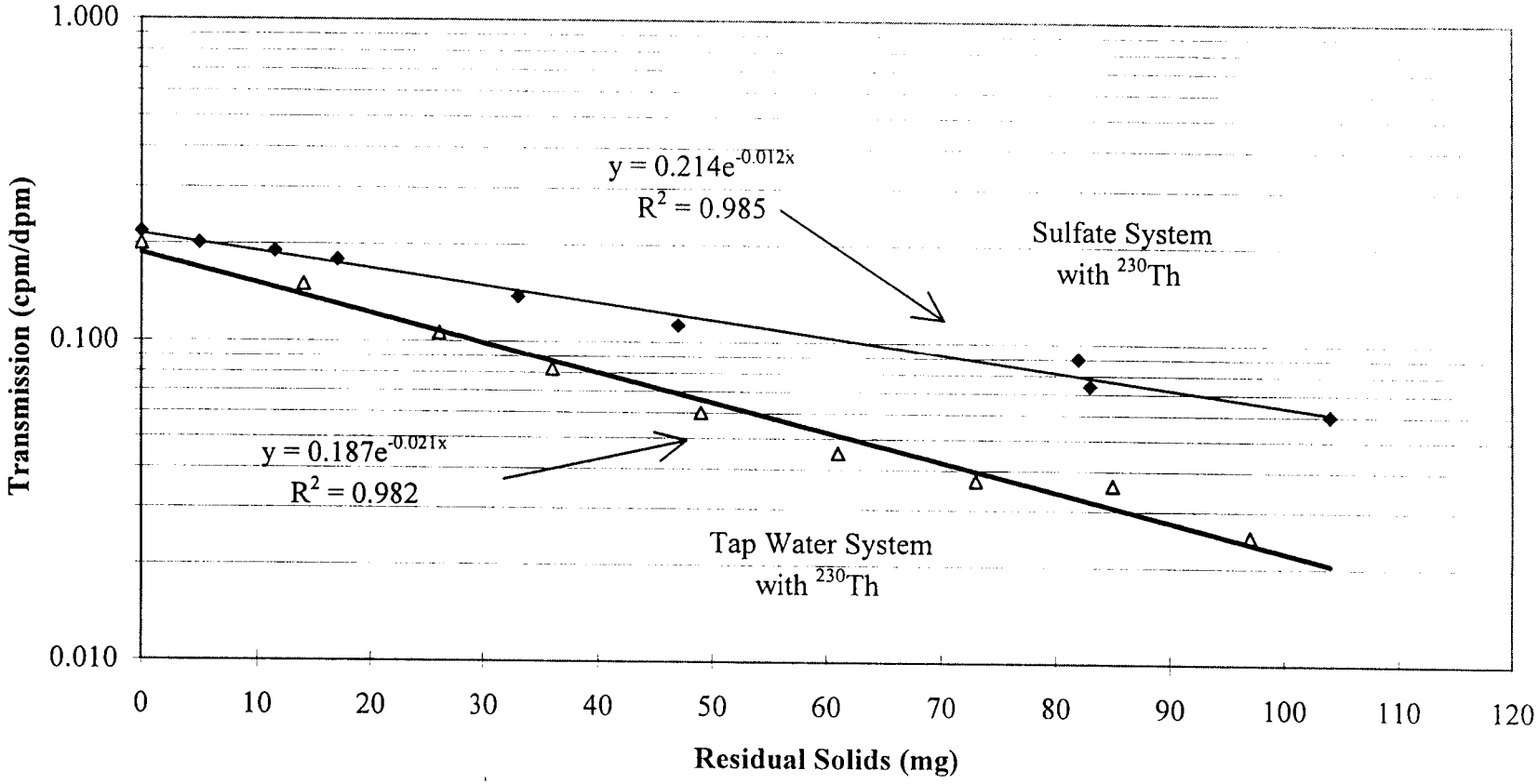

Figure 6. ^{230}Th Comparison

Figure 6. is included to demonstrate the different attenuation curves created from the EPA study using the same isotope. The upper curve included a sulfate salt solution to vary the water solids and the lower curve used tap water additions introducing nitrates to the samples. The EPA study fit third order equations to the data rather than the exponential fits shown in Figure 6. because it was more convenient for their software and because other groups used the same type of fit. An exponential fit presented here with EPA's data more closely resembles the type of curves normally found where attenuation and self-absorption are present.

4.5 ALPHA AMPLIFICATION FACTORS

As the voltage is increased on a proportional counter from the alpha to beta plateau, the number of alpha counts increases. If the detector does not discriminate for higher energy alpha pulses at the beta plateau, the amplified alpha activity at the beta plateau voltage must be subtracted from the total beta plateau count.

4.5.1 Plateau Curve Extrapolation

The positive slope at the alpha plateau attributes to the increased number of alpha counts at the beta plateau. By extrapolating a line tangent to the alpha plateau out to the beta operating voltage, a ratio describing alpha amplification from beta plateau to alpha plateau is found. The equation of the good linear fit shown in Figure 7. has a slope of 0.711 counts per volt, resulting in the calculated beta to alpha ratio of 1.56.

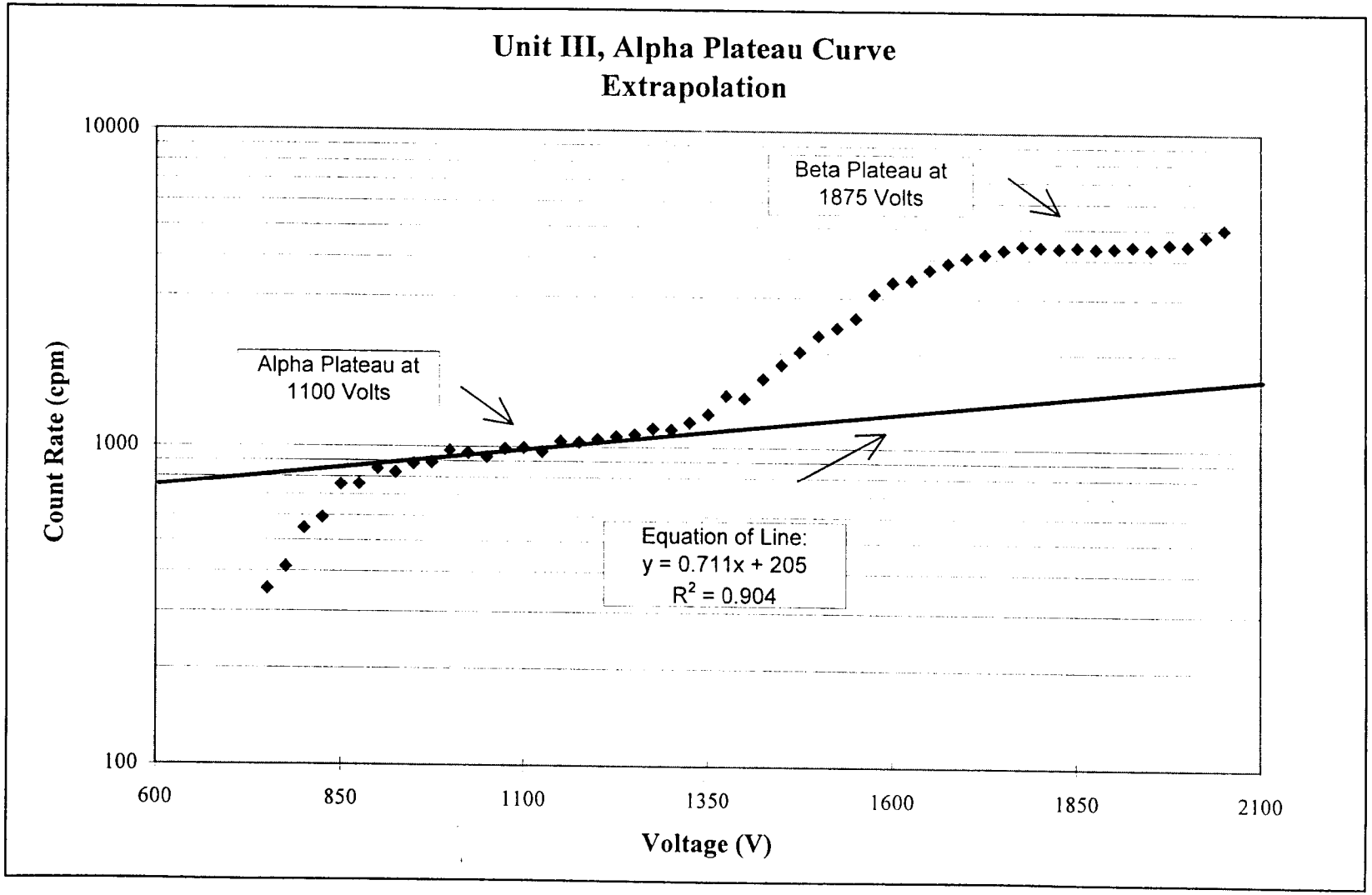

Figure 7. Unit III, Alpha Plateau Curve Extrapolation

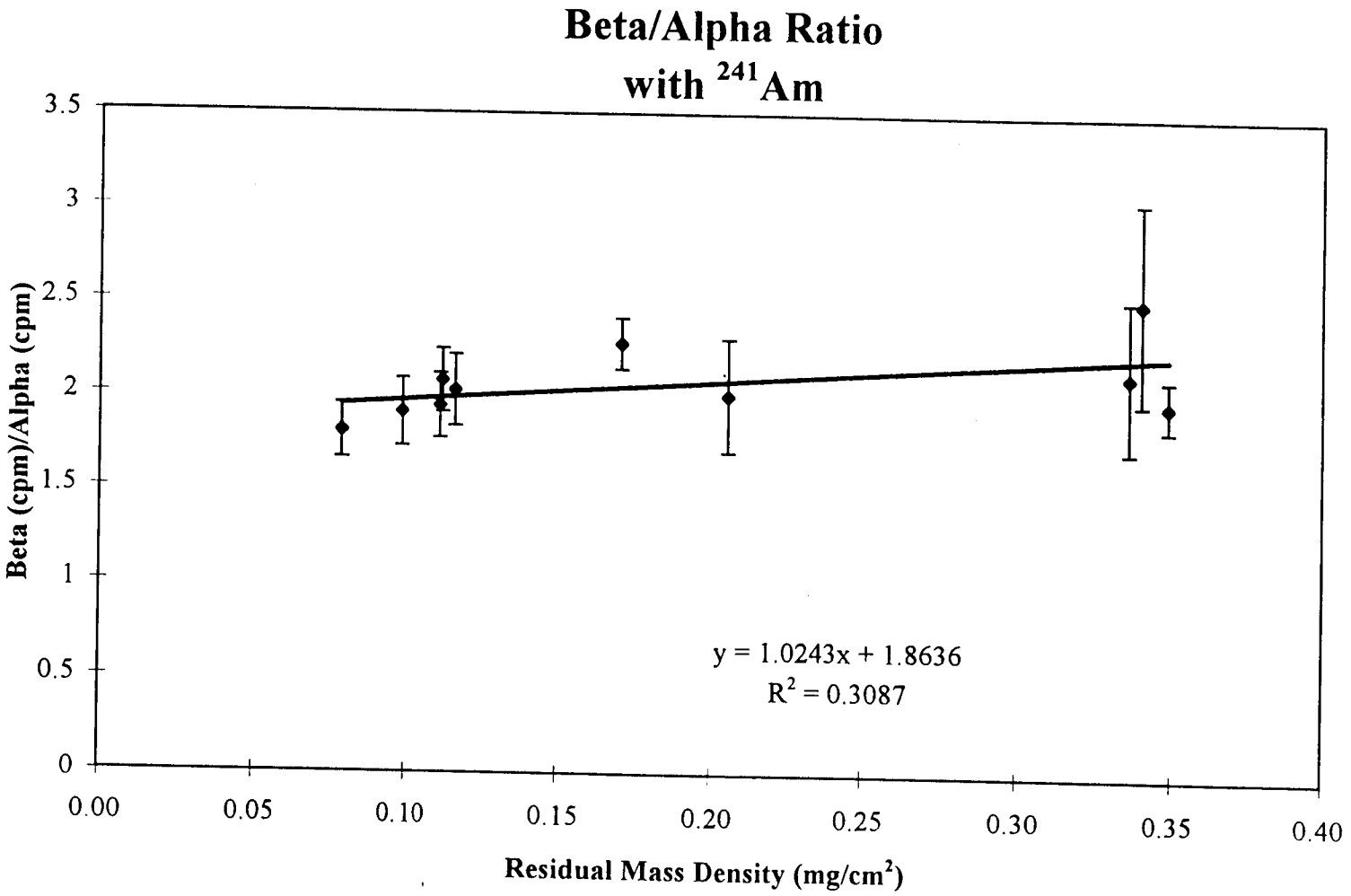

Figure 8. Measured Beta to Alpha Ratio

4.5.2 Measured Alpha Amplification

If a biosolids sample contains a mixture of both alpha and beta emitting nuclides, it is not valid to count the sample at the beta plateau and simply subtract the number of counts measured at the alpha plateau. The alpha amplification factor must be used to find the increased number of alpha counts at the beta plateau. Of course if a sample has no alpha component measured at the alpha plateau, the amplification factor is unnecessary.

The ^{241}Am planchets were counted at the beta plateau voltage to test the amplification factor. Figure 8. plots the number of measured counts at the beta voltage divided by the alpha counts (beta to alpha ratio) versus density thickness. The very poor fit to the data points shows a beta to alpha ratio of 1.86 at zero density thickness, but suggests problems. Explanations for this error may include inconsistent counting at either plateau, poor chemistry or the presence of a 60 keV ^{241}Am gamma or a host of other emitted particles. Assuming a zero density thickness allows a comparison of the beta to alpha ratio, but also includes a small amount of error as well. Finally, the error bars shown in Figure 8. comprise the error incorporated into each step of the sample preparation and counting process using error propagation techniques with 95% confidence.

4.6 GROSS ALPHA AND BETA TEST RESULTS

A one liter preserved test sample which originated from the CWRP sludge digester was analyzed. All subsequent gross alpha and beta tests came from this single sample. Ten planchets each with one milliliter of unspiked test biosolids were prepared by

Sample	Mass (mg)	Gross Counts per Minute	Background Counts per Minute	Net Counts per Minute	Net Disintegrations per Minute	Minimum Detectable Activity (dpm)
S-1	16.3+/-0.5	0.2+/-0.14	0.4+/-0.14	0+/-0.20	0+/-0.98	1.81
S-2	13.5+/-0.5	0.3+/-0.17	0.4+/-0.14	0+/-0.22	0+/-1.04	1.71
S-3	10.4+/-0.5	0.2+/-0.14	0.4+/-0.14	0+/-0.20	0+/-0.88	1.61
S-4	14.6+/-0.5	0.3+/-0.17	0.4+/-0.14	0+/-0.22	0+/-1.06	1.75
S-5	15.5+/-0.5	0.2+/-0.14	0.4+/-0.14	0+/-0.20	0+/-0.97	1.78
S-6	12.7+/-0.5	0.2+/-0.14	0.4+/-0.14	0+/-0.20	0+/-0.92	1.69
S-7	13.9+/-0.5	0.2+/-0.14	0.4+/-0.14	0+/-0.20	0+/-0.94	1.72
S-8	14.6+/-0.5	0.2+/-0.14	0.4+/-0.14	0+/-0.20	0+/-0.95	1.75
S-9	10.2+/-0.5	0.3+/-0.17	0.4+/-0.14	0+/-0.22	0+/-0.98	1.61
S-10	9.2+/-0.5	0.4+/-0.20	0.4+/-0.14	0+/-0.24	0+/-1.06	1.58
Sample Average	13.1+/-1.58	0.3+/-0.5	0.4+/-0.44	0+/-0.67	0+/-3.1	2.35

Table 6. Gross Alpha Test Results For Unspiked Sample

following the procedures listed in Chapter 3.4, Gross Alpha and Beta Determination. After weighing the samples and performing a quality control check on the Unit III GFPC, each planchet was counted for 10 minutes. Table 6. and Table 7. show the results. Not one measured sample contained above background levels of alpha activity, averaging 0.0 +/- 1.4 pCi/ml. The same planchets were then tested for gross beta and showed only planchet

Sample	Mass (mg)	Gross Counts per Minute	Background Counts per Minute	Net Counts per Minute	Net Disintegrations per Minute	Minimum Detectable Activity (dpm)
S-1	16.3+/-0.5	42.2+/-2.05	49+/-1.6	0+/-2.6	0+/-5.4	2.15
S-2	13.5+/-0.5	44.9+/-2.12	49+/-1.6	0+/-2.7	0+/-5.4	2.13
S-3	10.4+/-0.5	46.3+/-2.15	49+/-1.6	0+/-2.7	0+/-5.4	2.11
S-4	14.6+/-0.5	45.9+/-2.14	49+/-1.6	0+/-2.7	0+/-5.5	2.14
S-5	15.5+/-0.5	45.8+/-2.14	49+/-1.6	0+/-2.7	0+/-5.5	2.15
S-6	12.7+/-0.5	46.8+/-2.16	49+/-1.6	0+/-2.7	0+/-5.5	2.13
S-7	13.9+/-0.5	48.6+/-2.20	49+/-1.6	0+/-2.7	0+/-5.6	2.13
S-8	14.6+/-0.5	52.0+/-2.28	49+/-1.6	3.0+/-2.8	6.1+/-5.7	2.14
S-9	10.2+/-0.5	41.8+/-2.04	49+/-1.6	0+/-2.6	0+/-5.2	2.11
S-10	9.2+/-0.5	45.2+/-2.13	49+/-1.6	0+/-2.7	0+/-5.4	2.10
Sludge Sample	13.1+/-1.58	45.9+/-6.78	49+/-5.1	0.3+/-8.5	0.61+/-17	5.06

Table 7. Gross Beta Test Results For Unspiked Sample

S-8 to be very slightly above background. This behavior is most likely from detector abnormalities rather than any attributable radioactivity. The averaged beta activity measured was 0.3 +/- 7.7 pCi/ml. Ultimately, the values calculated for alpha and beta activity and minimum detectable activities incorporate attenuation effects and detector efficiencies by consulting the alpha and beta transmission curves. These results were compared to CWRP biosolids measured by CORE Laboratories in Casper, WY during

June, 1991. No statistically significant gross alpha or beta content was found by CORE, which is in agreement with the results obtained here. Before the investigation, it was expected that no gross alpha or beta contamination would be uncovered for Corvallis generated biosolids because the number of licensees contributing to the plant is relatively small. However, larger Oregon cities such as Eugene, Salem, or Portland have a higher probability of reconcentrating radioactive materials in biosolids due to the greater amount of licensees.

4.7 GAMMA SPECTROSCOPY TEST RESULTS

A gamma spectrum of preserved CWRP sludge was produced and analyzed with the Maestro II emulation program from EG&G Ortec using the procedures from section 3.6, Gamma Component Evaluation . The 80,000 second background count contained several small peaks originating from naturally occurring isotopes and possibly slight contamination in the cave. Once these background peaks were subtracted from the biosolids sample spectrum, two small peaks remained at 0.3638 MeV and 0.4769 MeV.

^{131}I

It is believed the 0.3638 MeV peak is a result from the ^{131}I gamma emitted with 81.2% probability. ^{131}I decays by the emission of several other gammas with much lower probabilities but no peaks above background were observed at the proper energies. Still, the possibility of observing ^{131}I was expected for two reasons. Firstly, Good Samaritan Hospital is licensed to use ^{131}I for therapeutic purposes, and its location is within a few miles from the CWRP. Plant operators estimate the time it takes for the hospital waste to

travel and be processed by the plant is two to three days. Secondly, because the sample was taken from the sludge digester rather than a lagoon, the sampling time was within the first 8.04 day ^{131}I half life and a higher concentration was expected.

An ADCAM 3 lithium-drifted germanium detector efficiency was determined from the Amersham Marinelli standard ^{57}Co and ^{137}Cs sources. Unfortunately, the isotopes with energies similar to the detected peak energy had decayed to background, therefore an interpolation using ^{57}Co and ^{137}Cs efficiencies of 7.68% and 0.78%, respectively, was used. The germanium detector efficiency for ^{131}I was 4.6%. Summing the peak area above the base line continuum gave 500 counts in an 80,000 seconds. With this information, an activity concentration was found to be 0.11 pCi/l +/- 0.01 pCi/l. However, the calculated MDA was 0.91 pCi; proving the measured concentration not statistically valid. (Assuming this activity concentration value is correct, and the sludge digester contains nearly one million gallons, the total activity in the digester at the time of sampling could have been 0.4 +/- 0.04 μCi of ^{131}I.)

^{7}Be

By process of elimination, the 0.477 MeV peak was thought to be caused by the ^{7}Be gamma emitted with 10.4% probability. All other possible reasons for this peak such as a double escape peak from other isotopes were checked and discarded. Consequently, further research presented answers for the detection of ^{7}Be. The National Council on Radiation Protection and Measurements (NCRP) Report No. 45, Natural Background Radiation in the United States discusses the origins and behaviors of ^{7}Be. (NCRP 45, 1975)

According to NCRP 45, an atmospheric production of ^{7}Be occurs through spallation of ^{16}O, ^{12}C, and ^{14}N, with an average cross section of 10 millibarns. Its production rates vary considerably with latitude and altitude, but remain fairly constant with time. Seventy percent of ^{7}Be production occurs in the stratosphere with the other thirty percent in the troposphere and atmospheric concentration levels have been shown to drop from rainout, washout and jet stream changes. A study at the Olympic Peninsula, WA, March 21, 1967, reported an average concentration of 26 pCi/l of ^{7}Be in rainwater. ^{7}Be also undergoes seasonal variations. A study in Richland, WA showed maximum concentrations in air during the spring and minimum concentrations during the fall and winter. Several studies have also shown the uptake of ^{7}Be in moss and lichen. Flora radionuclide concentrations in the state of Washington, Quinault rain forest, observed 30 μCi/kg wet weight in lichen during heavy rainstorms.

Comparing these findings to the conditions in Corvallis, OR, on May 21, 1996 reveals similarities. Samples were taken during the springtime, and rainfall throughout the month of May was 3.98 inches, more than twice the May average of 1.95 inches. (Taylor, 1996) On average, the sludge digester retains biosolids for 30 days, which coincides with the extremely wet spring weather. Examining these conditions show the detection of ^{7}Be in digested sludge during the rainy spring months may be reasonable.

The *Environmental Radiological Surveillance Report on Oregon Surface Waters* also mentions the presence of ^{7}Be in Oregon sediments. For the 1983 to 1993 time period, Willamette River sediments at Springfield and Harrisburg contained less than 0.1

and 0.7 pCi/g of ^{7}Be respectively, at a the two sigma level. (Oregon Health Division, 1994)

The preserved sludge sample gamma spectrum recorded 365 +/- 30 gammas during the 80,000 second counting time above the base line continuum. Assuming a detector efficiency interpolated from the ^{57}Co and ^{137}Cs sources to be 3.1%, the calculated concentration of ^{7}Be was 0.96 +/- 0.08 pCi/l. The calculated MDA for ^{7}Be was 1.1 pCi, which exceeds the measured activity concentration.

4.8 FURTHER RESEARCH

The logical progression of this inquiry regarding contaminated biosolids is the application to dose analysis scenarios. With the disposal locations for CWRP sludge determined, a full characterization of each field would allow accurate public dose assessments. The results could then be extended to set sludge concentration limits which would restrict its placement onto agricultural fields for the state of Oregon. However the steps for measuring the general radioactive material concentrations developed here are the required foundations for such extensions.

Throughout the investigation a few questions regarding equipment characteristics arose which could lead to further inquiry, such as the discrepancies and unusual results regarding proportional counter behavior. Why did the counts decline as a single sample planchet was repeatedly counted? Every factor seemingly possible, from voltage changes to improper purging, was checked and discounted, although the outcome on three separate detectors remained irregular. One possible reason could have been the high

activity associated with the metal planchets causing a static charge that may have slightly altered the electric field within the counting chamber. No matter the reason, the NMC proportional counters at the Radiation Center are growing obsolete and somewhat unreliable.

The proportional counter alpha amplification factor theory alone could provide a full inquest. For this work, the non-zero alpha plateau slope was produced using an assumed pure alpha emitting ^{241}Am source. In reality, the emitted 60 keV gamma contributed to the number of counts recorded at the beta plateau. This component could be accurately found by implementing a sample cover to attenuate the alpha while simultaneously unaffecting the gamma.

5. CONCLUSIONS

Using the general drinking water procedures established by the Environmental Protection Agency as a basis, an investigation of radioactive components in the local wastewater reclamation plant biosolids has been performed. This study has shown that the contamination of biosolids has occurred across the United States and that presently federal regulations regarding the sanitary sewer disposal of isotopes is in a state of flux.

General procedures have been created to determine the gross alpha, gross beta and gamma radionuclide concentrations in sludge samples. Supplementing the procedures, separate alpha and beta transmission curves have been developed to account for detector efficiency and source attenuation factors. An alpha amplification factor graph has also been produced to allow proper counting measurements taken at the GFPC beta plateau for mixed alpha and beta samples. By consulting the curves and following the procedures, 0.0 +/- 1.4 pCi/ml of gross alpha and 0.3 +/- 7.7 pCi/ml of gross beta have been measured in the local Corvallis Wastewater Reclamation Plant digested sludge.

A gamma spectroscopy analysis was also performed according to procedures and showed 0.11 +/- 0.01 pCi/l of ^{131}I and 0.96 +/- 0.08 pCi/l of ^{7}Be in Corvallis Wastewater Reclamation Plant digested sludge.

Even though future government regulations concerning radioactive materials in biosolids remain uncertain, Oregon State University and the state of Oregon will be prepared for any changes that may come.

BIBLIOGRAPHY

10 CFR 20. 1995. United States Nuclear Regulatory Commission. "Standards for Protection Against Radiation." Code of Federal Regulations.

40 CFR 141. 1995. United States Environmental Protection Agency. "National Primary Drinking Water Regulations." Code of Federal Regulations.

Ainsworth, C., R. Hill, K. Cantrell, D. Kaplan, M. Norton, R. Aaberg, E. Stetar. 1994. "Reconcentration of Radioactive Material Released to Sanitary Sewers in Accordance with 10 CFR 20." NUREG/CR-6289 (PNL-10193), Pacific Northwest Laboratory, Richland, Washington, Performance Technology Group, Inc., Nashville, Tennessee.

Agenda of Regulations. 1996. Nuclear Licensing Reports, PRM-20-22, Rockville, Maryland.

Binney, Stephen. 1995. NE/RHP 537. *Applications of Nuclear Techniques*. Oregon State University. Class Notes.

The City, 1995. "Combined Sewer Overflow (CSO) Remediation Plan Alternative Approved By Council." City of Corvallis, OR, Vol. 8, No. 12

City of Corvallis, Municipal Wastewater Reclamation Plant, 1978. City of Corvallis, OR, brochure.

Clark, Mike. 1996. Corvallis Wastewater Reclamation Plant Operator, personal communication. March 1, 1996.

Combined Sewer Overflow, Recommended Alternative. 1995a. November 6, 1995, City of Corvallis, OR, leaflet.

Combined Sewer Overflow, Choices and Alternatives. 1995b. September 1995, City of Corvallis, OR, leaflet.

Federal Register. 1986. Proposed Rules, Vol. 51, No. 6, 1116.

Federal Register. 1991. Rules and Regulations, Vol. 56, No. 98, 23361.

Federal Register. 1993. Proposed Rules, Vol. 58, No. 201, 54071.

Federal Register. 1994a. Proposed Rules, Vol. 59, No. 38, 9146.

Federal Register. 1994b. Notice of Availability, 64438.

GAO Report. 1994. "Nuclear Regulation: Action Needed to Control Radioactive Contamination at Sewage Treatment Plants." GAO/RCED-94-133, Gaithersburg, Maryland.

Halsey, R. 1986. "Investigation of Sewage System Contamination Incidents in Tennessee." State of Tennessee, Department of Public Health, Division of Radiological Health, Nashville, Tennessee.

Kennedy, Jr., W., M. Parkhurst, R. Aaberg, K. Rhoads, R. Hill, J. Martin. 1992. "Evaluation of Exposure Pathways to Man from Disposal of Radioactive Materials into Sanitary Sewer Systems." NUREG/CR-5814 (PNL-7892), Pacific Northwest Laboratory, Richland, Washington.

Knoll, Glenn F. 1989. *Radiation Detection and Measurement, Second Edition*. John Wiley & Sons, New York.

Krieger, H., E. Whittaker. 1980. "Prescribed Procedures for Measurement of Radioactivity in Drinking Water." EPA-600/4-80-032. Radiological Methods Section, Environmental Monitoring and Support Laboratory, Cincinnati, OH.

Larsen, I.L. 1995. "In-House Screening for Radioactive Sludge at a Municipal Wastewater Treatment Plant." *Radiation Protection Management*, Vol. 12, No. 4:29-38.

Lester, J.N. 1987. "Chapter 1, Primary Mechanical Treatment," pp. 1-14. *Heavy Metals in Wastewater and Sludge Treatment Processes, Vol. 2: Treatment and Disposal*, ed. J.N. Lester. CRC Press, Inc., Boca Raton, Florida.

Lide, David R. Editor-in-chief. 1994. *CRC Handbook of Chemistry and Physics*, 74th ed. 1993-1994. CRC Press, Inc., Boca Raton, Florida.

MacClennan, P. 1984. "4 Workers Get Clean Bill in Radiation Tests." *Buffalo News*, Buffalo, New York.

Marino, Gigi. 1994. "The Nuclides in Town, Does Danger Lurk in Low-level Radioactivity in Sewage?" *Science News* 146:218-219.

Metcalf and Eddy, Inc. 1991. *Wastewater Engineering, Treatment, Disposal, Reuse, Third Edition*. McGraw-Hill Publishing Company, New York.

Nuclear News. 1994. "Sewer Claims Cobalt-60 Threat." 37:67-68.

NCRP. 1975. *Natural Background Radiation in the United States*. Report No. 45. National Council on Radiation Protection and Measurements. Washington, DC.

Oregon Health Division, Radiation Protection Services. 1994. Environmental Radiological Surveillance Report on Oregon Surface Waters, 1963-1993. Volume I, December, 1994.

Parrotta, M. J. 1991. "Radioactivity in Water Treatment Wastes: A USEPA Perspective." *Journal of American Water Works Association*. April

Prewitt, J., and S. Glass. 1994. "Is Your Wastewater Radioactive?" *Water Environment and Technology*. 6:54-58.

Rimawi, K. 1984. "Americium-241 Contamination in Tonawanda and Grand Island, New York." State of New York, Department of Health, Bureau of Environmental Radiation Protection, Albany, New York.

Salvato, J. *Environmental Engineering and Sanitation*. 4th ed. John Wiley & Sons, Inc., New York.

Stetar, E, H. Boston, I. Larsen, M. Mobley. 1993. "The Removal of Radioactive Cobalt, Cesium, and Iodine in a Conventional Municipal Wastewater Treatment Plant." *Water Environment Research*, Vol. 65, No. 5, 630-639.

Taylor, George. 1996. State of Oregon Climatologist, personal communication. June 15, 1996.

Tchobanoglous, G., and E. D. Schroeder. 1987. *Water Quality, Characteristics, Modeling, Modification*. Addison-Wesley Publishing Company, Menlo Park, California.

US Environmental Protection Agency, (EPA). 1995. *A note regarding the Gross Alpha Portion of the October 27, 1995 Gross Alpha-Beta in Water Performance Evaluation Study*. National Exposure Research Laboratory, Office of Research and Development. Las Vegas, NV.

US Environmental Protection Agency, (EPA). 1989. "Development of Risk Assessment Methodology for Land Application and Distribution and Marketing of Municipal Sludge." Report No. EPA/600/6-09/001. Environmental Criteria and Assessments Office, Cincinnati, OH.

US Environmental Protection Agency, (EPA). 1986. "Gross Alpha and Gross Beta." Method 9310, Revision 0. SW-846, Ch. 6, Vol. IC. 54 FR 40263.

US Nuclear Regulatory Commission (NRC). 1986a. "NRC Region I Inspection of INS, Corp., Royersford, PA." Report No. 030-20934/85-04, King of Prussia, Pennsylvania.

US Nuclear Regulatory Commission (NRC). 1986b. "NRC Region I Inspection of Department of Health and Human Services, Rockville, MD." Inspection No. 86-01, King of Prussia, Pennsylvania.

Weekly Information Report. 1996. Week ending March 15, 1996, Nuclear Materials Safety and Safeguards, Enclosure B. SECY 96-061.

Wheatley, Andrew, editor. 1990. *Anaerobic Digestion: A Waste Treatment Technology.* Critical Reports on Applied Chemistry, Vol. 31. Elsevier Applied Science, New York.

Zeyher, Allen. 1995. "Sewage Treatment Plants Need to Assess Radioactivity." *Nuclear News.* 38:55-56.

APPENDICES

Appendix 1.

10 CFR 20.2003 (1-1-95 Edition)
Disposal by Release Into Sanitary Sewerage.

§ 20.2003 Disposal by release into sanitary sewerage.

(a) A licensee may discharge licensed material into sanitary sewerage if each of the following conditions is satisfied:

(1) The material is readily soluble (or is readily dispersible biological material) in water; and

(2) The quantity of licensed or other radioactive material that the licensee releases into the sewer in 1 month divided by the average monthly volume of water released into the sewer by the licensee does not exceed the concentration listed in table 3 of appendix B to §§20.1001-20.2401; and

(3) If more than one radionuclide is released, the following conditions must also be satisfied:

(i) The licensee shall determine the fraction of the limit in table 3 of appendix B to §§20.1001-20.2401 represented by discharges into sanitary sewerage by dividing the actual monthly average concentration of each radionuclide released by the licensee into the sewer by the concentration of that radionuclide listed in table 3 of appendix B to §§20.1001-20.2401; and

(ii) The sum of the fractions for each radionuclide required by paragraph (a)(3)(i) of this section does not exceed unity; and

(4) The total quantity of licensed and other radioactive material that the licensee releases into the sanitary sewerage system in a year does not exceed 5 curies (185 GBq) of hydrogen-3, 1 curie (37 GBq) of carbon-14, and 1 curie (37 GBq) of all other radioactive materials combined.

(b) Excreta from individuals undergoing medical diagnosis or therapy with radioactive materials are not subject to the limitations contained in paragraph (a) of this section.

Appendix 2.

Selected Listings of 10 CFR 20 Appendix B, Table 3
Average Monthly Concentrations
Allowed for Sanitary Sewer Release

Atomic Number	Radionuclide	Monthly Average Concentration (μCi/ml)
1	Hydrogen-3	1E-2
4	Beryllium-7	6E-3
5	Carbon-14	3E-4
11	Sodium-24	5E-4
15	Phosphorus-32	9E-5
16	Sulfur-35	1E-3
17	Chlorine-36	2E-4
19	Potassium-40	4E-5
24	Chromium-48	8E-4
27	Cobalt-60	3E-5
31	Gallium-67	1E-3
38	Strontium-89	8E-5
38	Strontium-90	5E-6
39	Yttrium-90	7E-5
43	Technetium-99m	1E-2
43	Technetium-99	6E-4
53	Iodine-123	1E-3
53	Iodine-125	2E-5
53	Iodine-131	1E-5
55	Cesium-137	1E-5
81	Thallium-201	2E-3
82	Lead-210	1E-7
88	Radium-226	6E-7
92	Uranium-238	3E-6
94	Plutonium-239	2E-7
95	Americium-241	2E-7