Fate and mobility of radium-226 in municipal wastewater sludge following agricultural landspreading: a survey. [DNR-019] [1987]

Portle, Thomas L.
Madison, Wisconsin: Wisconsin Department of Natural Resources, [1987]

https://digital.library.wisc.edu/1711.dl/DLLU7VJBBFZG684

Wisconsin Groundwater Management Practice Monitoring Project
No. 22

GROUNDWATER
Wisconsin's
buried treasure

Wisconsin Department of Natural Resources

Fate and Mobility of Radium-226 in Municipal Wastewater Sludge Following Agricultural Landspreading

A Survey

Wisconsin Department of Natural Resources
Box 7921, Madison, WI 53707

PUBL-WW-006 87

Prepared By

Thomas L. Portle

ACKNOWLEDGMENT

The author wishes to thank the following people who provided invaluable assistance in the production of this report:

Bureau of Wastewater Management District and Central Office Staff as well as Treatment Plant Operators were very helpful in the selection and investigation of municipalities and sludge landspreading application sites. Special thanks is due to Tom Nelson, Sludge Management Specialist in the Southeast District, and Bob Schaefer, former Engineer in the Southern District.

Appreciation is expressed to Carolyn Hunger, Bureau of Wastewater Management, LTE, for her thorough field and office efforts in data acquisition, sample handling, transport and recordkeeping, making contacts with and follow-up correspondence to land owners and treatment plant officials.

Diane Brinson, Bureau of Information and Education and Cindy Lukas, Bureau of Wastewater Management provided critical editorial review which greatly enhanced this document.

DNR staff who commented on the various drafts of this document.

Pat Parsons, Department Librarian for her diligence in tracking down much of literature cited including some very obscure publications.

Dave Schliss, of the Laboratory of Hygiene, performed rapid sample analysis and obtained necessary reference material to install laboratory procedures not previously available, but critical to the completion of this report.

The Second Floor Word Processing staff for their professionalism and timeliness.

Photo Credits

Thomas L. Portle: Cover

9351V

State of Wisconsin \ DEPARTMENT OF NATURAL RESOURCES

Carroll D. Besadny
Secretary

BOX 7921
MADISON, WISCONSIN 53707

July 24, 1987

IN REPLY REFER TO: 3420

Interested Community Officials,
Consulting Engineers,
Treatment Plant Operators,
Manufacturers, &
Regulatory Agencies

The purpose of this report is to provide background information regarding radium-226 in municipal wastewater sludge and alert you to the potential that its presence may affect sludge disposal.

The presence of radium-226 and its decay product, radon gas, in the environment is of growing concern. Radium is found in soils, in rock formations and in groundwater. Increasingly, radium and radon gas are being acknowledged as health hazards. If groundwater is in contact with certain rock types, then radium in the groundwater can be present at unacceptable levels. When wastewater sludge is in contact with such water it adsorbs radium. For these reasons the attention of the Department of Natural Resources has been focused on radium.

The Bureau of Water Supply routinely sampled water sources across the state and found that in 46 out of approximately 1,300 communities the combined radium level exceeded the state and federal drinking water standard of 5 picocuries per liter (pCi/l). Having learned this, the Bureau of Wastewater undertook a study to determine if radium from the water supply could become concentrated in wastewater sludge. This study established that wastewater sludge concentrated radium.

Radium-226 presents a potential hazard when ingested. While there is a drinking water standard for radium-226, little was known about the fate of radium-226 in sludge after it has been applied to agricultural fields. This led to a decision to gather more information about the safety of land application of sludge from these communities on agricultural lands.

A field study was designed to determine if this practice constitutes a threat to food chain crops or to the groundwater. We studied how conditions at the sludge application sites affect radium mobility and behavior. Samples of common agricultural plants grown on the soils were collected to see if evidence of plant uptake could be found. Also, our data on radium levels in soils, plants and sludge were compared to the literature and to common agricultural fertilizers to gain a perspective on the potential hazard.

The results of the study are the subject of this report and were used to draft guidelines to safely manage the landspreading of sludge which contains radium. These guidelines have been approved by the Radiation Protection Council.

Future plans are to sample selected communities for radium in sludge and to determine how widespread the problem is. All those communities with a violation of the drinking water standard will be sampled if they produce a sludge which is landspread. Also, there are cases where communities whose water supplies do not exceed the radium drinking water standards could still result in sludge radium levels that exceed the level of concern (2 pCi/gram dry weight). Therefore, sampling needs to be conducted in all localities where a problem may exist.

In cases where a sludge exceeds the action level, permittees will receive further information on how to proceed. This will include guidelines for the safe disposal of sludge that contains radium-226 which were generated in this report. When warranted, there will be monitoring requirements in future WPDES permits. Additional data will be generated through adherence to the monitoring requirements contained in the landspreading guidelines. These data will provide the basis for any future adjustments in the guidelines which may be warranted.

While the data base is not yet available to provide final confirmation, it appears that the landspreading of sludge containing radium may continue under these guidelines without threat to the environment.

Sincerely,

CD Besadny

C. D. Besadny
Secretary

9351V

Fate and Mobility of Radium-226 in
Municipal Wastewater Sludge Following Agricultural Landspreading

A Survey
PUBL-WW-006 87
July 1987

TABLE OF CONTENTS

LIST OF TABLES

7145V

EXECUTIVE SUMMARY

A field study was conducted to evaluate the fate of radium-226 applied incidentally during the landspreading of municipal sludge. Survey results were compared to data available in the literative and were used to develop policy. Those communities in which a violation of the combined radium drinking water standard existed, and which produced a sludge that is landspread were sought. For selected communities, sludge spreading sites and corresponding control sites were chosen based on their sludge application history and the soil properties. Composite soil samples for both the plow layer and the soil layer immediately below it were obtained. When possible, plant tissue was obtained and separated between the fruit or grain and the remaining above ground biomass.

Data indicated that soil radium activity levels were not elevated as a result of landspreading of sludge containing radium. Native radium levels appear to be related to clay content. No evidence that radium presented a threat to the groundwater was obtained. When soil radium activity levels in the lower soil increment exceeded those in the upper increment, clay levels in the lower increment were correspondingly higher.

The existence of an arcopetal gradient for radium in plants was supported. No elevation of radium activity in plant grain or fruit was observed. However, in some instances elevated radium was found in the above ground biomass in corn, soybeans and alfalfa. Plant radium activity levels did not appear to be directly related to soil radium activity levels. Plant radium levels were higher when plants were grown in soils which had lower clay contents and lower pH. This suggested that soil factors dictated plant uptake of radium.

Radium mobility is influenced by soil factors; this suggests that selection for and management of soil factors is a viable method of reducing radium mobility and minimizing environmental concerns. A set of guidelines to facilitate the safe land application of sludge contained in radium were developed and are included in this report.

9351V

I. INTRODUCTION

Background

The Wisconsin Departments of Health and Social Services and Natural Resources agreed in 1978 that the DNR Bureau of Water Supply would routinely sample drinking water in communities statewide. This sampling revealed that out of 1300 community water systems in Wisconsin, 44 exceeded the combined radium (radium-226 and radium-228) drinking water standard of 5 picocuries per liter (pCi/l) (Hahn, 1984). To determine the extent of radioactivity in Wisconsin groundwater, DNR conducted a study featuring selective well sampling. This study obtained broad information about radium in Wisconsin groundwater, gathered information about specific geologic formations, and investigated removal and disposal methods of radium (Hahn, 1984).

This study recommended that blending or use of alternative water supplies are the favored alternatives because disposal of solid waste products from radium removal processes could be expensive and complex. (Hahn, 1984).

The DNR study documented the extent of the radium contamination problem in the water supply. It prompted a subsequent study performed by the DNR's Bureau of Wastewater Management (in 1985) to ascertain the fate of radium in the wastewater treatment process. This study addressed the accumulation of radium in sewage sludge.

The study clearly demonstrated that sludge adsorbed radium wastewater. Even when the influent to the wastewater treatment plant contained low levels of radium, sludge concentrated the radium which accumulated either in the dissolved or the solid portion (Williams, 1985). Further, the recommendation was made that a subsequent study be performed to provide insight into the potential environmental impacts and hazards associated with the landspreading of a wastewater sludge containing radium. The survey discussed in this report was designed to address plant uptake and potential groundwater contamination from sludge landspreading. In addition, this survey will: 1) assist the Department in setting standards for radium sludge disposal, and 2) provide an adequate technical basis to assist decision making and the development of guidelines for the disposal of sludge containing radium.

II. OBJECTIVES

The objectives of this survey are to evaluate:

1. The mobility of radium within and through the soil profile;

2. The potential for groundwater contamination attributable to land application of sewage sludge containing radium;

3. The input of radium directly attributable to the land application of sewage sludge by obtaining the background radium-226 levels

found in similar local control sites which have not received sludge and comparing them with sites used for sludge disposal;

4. The factors related to sludge type, mode of application and/or relevant soil properties which are important in controlling radium mobility in soils;

5. The comparison of the radium levels in soil/sludge mixtures and plant samples obtained from sludge disposal sites in the municipal wastewater survey with levels found in various soils, plants, foods and other common materials on a nationwide and a worldwide basis;

6. The occurrence and magnitude of radium uptake and compartmentalization in various crops.

III. METHODS

A. Selection of Municipalities and Disposal Sites

Since our intent was to observe the most serious potential radium hazard in a landspreading system, it was important to locate communities with high radium levels in their water supply. High levels exceed the health standard of 5 pCi/l.

It was important that a community's water treatment processes not remove radium so that it would reach the wastewater treatment plant. Next, it was necessary that the community's wastewater treatment plant produced a sludge which was applied to agricultural lands.

The first step was to select communities: 1) which had a violation of the drinking water standard; and 2) which had a mechanical sewage treatment plant; and 3) either did not (a) treat drinking water, or (b) discharge backwash from treatment into the sanitary sewer.

Once the communities were selected the sludge spreading sites were identified. The soils at these sites offered the greatest probability of elevated radium levels attributable to landspreading thereby posing a potential environmental hazard. The following site selection criteria were employed:

1. History of wastewater sludge application;

2. Ability to allow observation of a range of soil properties and interactions with the sludge; and

3. Ability to portray a crop, sludge type, or a land application practice not available at other sites.

B. Sampling

To obtain the background radium activity levels in the soil, the Department selected control sites which had not received sludge containing radium (e.g. all radium activity was native or from other sources). These sites were of the same geologic origin as those which received sludge containing radium. All control sites were of the same soil series, general locality and land use history.

Soils were sampled at land application sites and at the control sites. At all sites, soils were sampled in the upper soil increment to a depth of 15 centimeters with a soil probe. Then a spade was used to create a 1-foot square space in the soil to the sample depth of the upper soil increment to prevent inadvertent contamination of the lower increment (Dr. Corey, 1985). The lower increment was sampled at the same location. A separate probe was used to obtain the sample of the lower 15 centimeter increment. Five or six probes (subsamples) were obtained. This constituted a composite sample and yielded a soil volume of at least 1,000 grams. Typically a "W" sampling pattern, but in some cases a straight line, was laid out on each site to obtain the necessary number of subsamples to yield a site-representative composite sample of adequate volume (Liegel, et al., 1980).

All equipment was thoroughly cleaned between sites to prevent cross contamination. Uniformity of sludge application was assumed.

When available, plant material was obtained as close as possible to the soil subsample site, with equal amounts being taken at each location to constitute a composite sample. The number of plant tissue samples was equal to the number of soil subsamples.

To determine if radium was taken up and subsequently translocated within plant specimens, plant tissue was separated between vegetative material (stalks, stems, leaves) and grain or fruit. Root samples were not obtained.

To determine the percentage of radium that was removed in the sludge, influent and effluent samples were taken from composite samplers at the wastewater treatment plants.

Sludge samples were taken at the point where it was normally removed and sampled to satisfy land application permit requirements.

C. <u>Laboratory</u>

Sample bags were labeled and taken to the Soil and Plant Analysis Lab for processing. The State Lab of Hygiene then analyzed the samples for radium-226 activity. After analysis, the sample was returned to the Soils and Plant Lab for appropriate agronomic analysis for pH, organic matter, cation exchange capacity, particle size, nitrogen, phosphorus, potassium, calcium, magnesium.

Briefly, methods for radiological analysis were: (for additional detail see Appendix II) 1) For water samples, radon-222 was sampled for alpha activity after an ingrowth period. Four pounds of plant material (wet-weight) were obtained. Samples were dried, ground and placed to yeild a 4 gram sample of ashed vegetation (in a muffle furnace). This was processed and after an ingrowth period of at least 14 days, gas (radon-222) was purged and counted for alpha activity. 2) For both soil and sludge testing 500 gram samples were processed and after a 28 day ingrowth period, gas was counted and gamma ray detection was employed to determine the radium-226 activity.

IV. RESULTS AND DISCUSSION

A. Influent, Effluent and Sludge Levels

Radium-226 activities found in influent, wastewater sludge and effluent for sampled communities are given in Table 1. Radium activity levels in influent are lower than water supply radium activity levels (as indicated by the Bureau of Water Supply radium violation list) in all cases. This difference can be explained as follows:

1. Radium was somehow removed as it passed through the homes and plumbing of individual consumers (possibly by removal in water softeners or tied-up in plumbing).

2. The radium-228 was not obtained for influent samples.

3. The inherent variability of water supply based on fluctuating use of various wells, which may represent different geologic formations (aquifers) and produce water of varying radium concentration.

4. Current remedial actions such as blending with alternative water sources by some of the communities in violation reduced the influent levels.

In each case, at least 60% of radium was retained in wastewater sludge (Table 1). This was expected since Williams (1985) found a) that wastewater treatment removed soluble radium from wastewater, b) biological sludge removed or adsorbed insoluble radium and, c) sludge treatment concentrated radium. This study was conducted in a similar water supply and sludge environment in Wisconsin.

Also, a study by Tsekos and Keller (1983), revealed that activated sludge was an excellent bio-adsorber of radium-226. One municipal sludge in that study adsorbed up to 75,000 pCi/gram under experimental conditions. Likewise, Rusanova (1964) indicated that organic matter, in general, has a large capacity to adsorb many elements and specifically accumulates radioactive substances.

The difference in radium retention between sludges (Table 1) may have to do with the contact time with the sludge, the volume of water involved, the radium activity of the water or some qualitative sludge difference which affected its adsorption ability. It appears that when higher influent was measured, higher retention was observed in the sludge and sludge radium-226 activity usually was higher (Table 1). This difference could be related, in part, to physical settling of insoluble radium. Williams (1985) alluded to physical settling as one means of radium removal.

Hazard Perspective

To provide some prospective on the hazard related to land application of sludge containing radium, the sludge-radium activity levels found in this study are presented in relationship to other sludges and common materials in Table 2.

It can be seen that sludges in this study (17.43 pCi/gram) do not show elevated radium activity when compared to common materials such as phosphate fertilizers (21 pCi/gram) or coal ash (24 pCi/gram). The elevation of soil radium activity levels as a result of use of phosphate fertilizers over an extended period would approximate the increase which results from the agricultural application of wastewater sludge containing radium. Hollins (1977) used the results of many worldwide studies to calculate permissible body burdens from plant foods. He calculated that if a garden were used for 50 years, the maximum permissible soil radium activity would be 220 pCi/g. The soils sampled in this survey did not exceed 1.78 pCi/g.

The hazard associated with emanation of radon daughters (gases produced during radium decay) can be compared to the federal acceptable standards for uranium tailings that radium-226 shall not exceed the background by: 1) 5 pCi/gram averaged over the first 15 centimeters of the soil, and 2) 15 pCi/g averaged over 15 cm thick layers of soil more than 15 cm below the surface (Federal Register, 1983). None of the soil/sludge mixtures from sites which had received sludge containing radium exceeded 1.78 pCi/gram. Because of well ventilated conditions and relatively low radium activity on sludge sites, no health hazard is anticipated.

B. Radium-226 Activity in Soil at Sludge Disposal Sites and at Corresponding Control Sites

To determine if the land application of sludges containing radium has caused an elevation of soil radioactivity, the soil radium activity at agricultural sites which received radium-containing sewage sludge is compared with corresponding control sites in Table 3. These results show that the application of sludge containing radium resulted in no apparent change in the site radium activity level. Indeed, in some cases such as Mayville, Mukwonago and Peshtigo, the control sites showed a higher radium activity than the sludge sites. As a result, soil radium levels may not be directly related to sludge landspreading. Both the average control site (0.81 pCi/gram) and average sludge site (0.91 pCi/gram) had radium activity levels which corresponded to the worldwide average soil level of 0.8 pCi/gram given by Osborne (1969). The 0.91 pCi/gram average sludge site level was much lower than average levels in western Europe and Great Britain, which are about 2 pCi/gram and 1.3 pCi/gram respectively; and much lower than extreme levels found in Iran, which can vary anywhere from 17 to 9,000 pCi/gram (Brooks, 1972).

1. Native Radium

The radium activity level present on any site in this survey could have been derived from parent material, the application of phosphate fertilizer, or the application of sludge-bearing radium. While the origin of the radium cannot be ascertained with certainty, some observations can be made.

It is highly likely that much of the radium is naturally occurring. Uranium is widely distributed in terrestrial environments, occuring in igneous and sedimentary rocks (Osborne, 1965). An equilibrium exists between uranium and radium (Mazor, 1963). Given the extensive glacial history of Wisconsin, it is likely to be found anywhere, including portions of the unglaciated area. Clay soils contain various amounts of primary minerals which have been weathered to clay size (Gillott, 1979); and clay often contains uranium and its daughter radium.

2. Phosphate Fertilization

Uranium and radium occur in the geologic formations which are mined for phosphate fertilizer. The mining and processing of these ores leaves uranium and radium in agricultural fertilizers.

The application of phosphate fertilizers contributes uranium and radium to the environment (Menzel, 1968; Guimond, 1978). The application of typical agronomic amounts of such fertilizer over a 50-year period can result in a build-up of 1 pCi/gram in the soil depending on the type of phosphate fertilizer used (Guimond, 1978). This generally agrees with research on heavily fertilized crops such as vegetables, tobacco, potatoes, and sugar beets. It was determined that additions of radium incidental to phosphate fertilization had approximately equaled the radium amounts which were naturally present in soils (Talibudeen, 1964).

3. Sludge Containing Radium

The amount of radium added to farmland through the land application of wastewater sludge is quite variable (Table 2). Factors contributing to this variability include: a) the radium level in the community's water supply, b) the municipality's efforts at blending or other treatment options to reduce radium levels in water supply, c) the sludge characteristics and contact time with the water supply. Calculations compared radium additions when sludge was applied to the nitrogen limit of the crop. These indicated that radium input to soils would typically add about 1 pCi/gram to the soil in an 18-20 year period. Although based on limited data, but indicates that in some instances sludge application could result in a more rapid radium elevation than that associated with the application of phosphate fertilizer.

C. Fates of Radium in the Environment

Soil radium, whether native or applied extraneously, is subject to the same set of possible fates. Radium can:

1. Remain in place, decay and dissipate in a gaseous state (emanation).

2. Leach downward in the soil profile, and may eventually reach groundwater.

3. Be taken up by plants.

4. Adhere to soil particles, and/or leave the site through surface erosion.

With any of the above pathways, there are potential environmental impacts and human health consequences. Of the above fates, the wastewater survey addressed plant uptake and leaching. Results of the survey are discussed below. Each fate, however, will be discussed briefly.

1. Emanation

As previously mentioned, radium and uranium are in equilibrium in soils (Mazor, 1963). Radium decays to radon gas. Because of the long half-life of radium-226 (1622 years), this decay is expected to continue indefinitely. The associated health hazard has received much attention. It has been estimated that indoor radon exposure may be linked to 30,000 deaths yearly in the United States (Environmental Pipeline, 1986.) Building foundations, basements, surrounding soils and building materials often contain radium. As homes have become better insulated, this threat has increased. In Tennessee, it is estimated that 1/3 of the homes may have unacceptable radon gas levels exceeding 0.02 working levels (background is 0.004 working levels).

In 1984, the Wisconsin Department of Health and Social Services, Section of Radiation Protection evaluated radiation risks to plant workers from radon gas at a drinking water treatment plant in Elkhorn, Wisconsin. The study concluded that an occupational or public health hazard did not exist due to radioactivity (Hahn, 1984).

2. Movement to Groundwater

The potential risk of groundwater contamination due to downward migration of radium was addressed in the wastewater survey. Results are given in Table 4. If radium activity in the lower increment of soil exceeds the radium activity of the upper increment of soil, this may be an indication that radium is moving downward in the soil profile.

Downward migration of radium was not evident in this survey. In cases where lower soil increment radium activity exceeded the upper increment on sludge sites, the clay content of the lower increment was greater than or equal to that of the upper increment. In this respect, radium activity was no different than the control sites where the same relationship was observed. It appears that native radium associated with the clay is important in accounting for differences between sites. In any case, there was no indication that radium, whether native or extraneously applied, migrated downward in the soil profile.

3. Retention by Clay

Clay has a moderating effect on the downward movement of cations, including radium, in the soil profile.

The literature agrees that a strong affinity exists between clay and radium. In one study, radium was more slowly released from clay soils than other similar alkaline earth cations such as calcium, barium and strontium (Menzel, 1968). Similarly, the authors found that the soil horizon which had lost its calcium due to leaching still retained radium (Telefair, 1960). This was attributed to the tenacity with which radium was held in clay soils.

Mechanism of Radium Retention by Clay

Since it appears that clay will adsorb and retain radium, it is necessary to understand the mechanism through which this is accomplished along with its limitations.

Clays have an ability to attract and retain ions due to their unsatisfied valences on inner and outer edges resulting from inbalances generated by isomorphic substitutions in the structure. Also, cations can be held in the space between clay layers. Clays are fine-sized particles (smaller than 0.002 mm), and often contain some primary minerals which are reduced to clay-sized range (Gillott, 1979). The quantity of negative exchange sites depends on the type of clay which affects the amount of surface area available for adsorbing cations. A 2:1 lattice clay, such as montmorillonite, typically has greater unsatisfied charge and surface area than a 1:1 clay such as illite. Any cation including radioactive ions can be adsorbed to the outer charge or held inside the lattice as a result of inner charge deficiencies. Thus, radium may be held like calcium or magnesium between plates or on the surfaces of plates. Radium held on the outer surface of plates is exchangeable to a larger extent than that held between the plates. In some instances, ions such as potassium are fixed (Schultz, 1965) and considered not available for further exchange.

4. Effect of Clay on Radium Mobility

Many authors have discussed the relationship of clay content and the immobilization of radium. Osborne (1965) in a review of the literature uncovered a number of significant relationships between radionuclides and soil types. First, the lattice alumino-silicate clays are efficient adsorbers of radium. Secondly, uranium (radium's parent) may be substituted for isomorphically. When (Delwiche, et al., 1958) measured the radon gas generated by various soils, those with the greatest clay content had the highest radon generating power. Radium was capable of being held in crystals and thus not available in soil solution.

It is evident that clay plays a major role in reducing radium mobility, and that it contains both native radium and applied radium. If the contribution of sludge applied radium is low when compared to total radium, you would expect to find little difference in control sites versus sites which had received sludge containing radium.

In this survey, a slightly higher average radium activity existed in the radium sludge sites versus control sites. There is no evidence that this difference was a result of sludge application. When all sludge sites and control sites are viewed together (Table 3), this can be seen readily. As soil texture became finer radium activity increased (Table 5). Indeed, an even more evident relationship exists between radium and clay content in this study. This agrees with the literature (Marple, 1980; Osborne, 1965).

The effect that clay exerts on radium mobility, as indicated by plant uptake of radium is important. This effect was apparent from the survey results. Plants grown in soils of low radium activity, but with low clay content (5%), had radium activities similar to those grown in soils with up to three times or more the radium activity (Table 7), but with high clay content (31 or 36%) clay.

Marple (1980) noted a similar effect when <u>Sitanion</u> was grown on uranium tailings. As percentage fines increased, the radium content of <u>Sitanion</u> decreased. She concluded that radium uptake was inversely related to texture.

In another study, Delwiche, et al. (1958) concluded that half the radon generating capacity was from radium "fixed" in the soil. Further, the amount of radium "fixed" in soil was directly related to clay content. Similarly, Marple (1980), on inactive uranium tailings, noted radium activity increased with fines content. Thus, it appears that clay content and its ability to attract and retain radium is critical in the control of plant uptake of radium. It must be remembered that there is a very limited number of observations in this survey, but the clay content of soil is nonetheless a worthwhile consideration in site selection. This tendency of clay to influence radium mobility and its practical application is in need of further research.

Management Implications

Sites selected for the application of radium-containing sewage sludge should possess as much clay as possible, balanced with the need to maintain adequate infiltration capacity. This will be further discussed in the guidance. This is a good opportunity for management since percentage fines and clay content are easily measured in the laboratory and observable in the field. Also, soil texture is documented in the county soil surveys. Proper site selection and management will result in a high probability of radium retention by adsorption, entrapment, or fixation mechanisms. In this way, leaching into groundwater or uptake by plants will be impeded to the extent that radium mobility is decreased.

5. Influence of Organic Matter on Radium Mobility

The affinity of organic matter for radium in wastewater treatment processes was established by Williams (1985). His data showed that wastewater sludge adsorbed soluble radium and alluded to its ability to obtain insoluble radium as well. Likewise, soil organic matter will, show an affinity for soluble radium since organic matter derived from sludge is similar to that found in soil. The interaction of soil organic matter with soluble radium, a divalent cation, is expected to be similar to its interaction with the metals (also divalent cations).

Soil organic matter has the ability to bind metals through its cation exchange capacity and as a result of the formation of organo-metallic complexes (Mortensen, 1963). The ability of organic fractions to bind metals has been well studied. Even so, in a recent paper it was stated that because of its complex nature, an adequate working knowledge is still not available (Logan and Chaney, 1983).

In one study, soil organic matter variation played a decisive role in controlling the mobility of radium and plant radium uptake in alfalfa and orchard grass in soils of similar radium activity. A seven-fold increase of radium activity in grass was observed when organic matter was 0.4 percent versus when it was 3.4 percent. Similarly, in alfalfa a four-fold increase was observed in the lower soil organic matter condition (Grzybowska, 1974).

For these reasons, soil organic matter content was given considered in the wastewater survey. Results are depicted in Table 6. It is evident that radium activity increased with organic matter content. Apart from the affinity of organic matter for radium, there is no explanation for this. It follows, however, that the selection of application sites which are relatively high in organic matter is prudent.

A percentage of the radium is adsorbed or retained in the organic matter as a result of wastewater treatment. This suggests that radium availability to plants could be dictated in part by the decomposition of the organic fraction to which it is bound. Even when radium is released due to the decomposition of organic matter, soil mechanisms are expected to act to minimize radium availability to plants.

Questions remain about the potential for chelation or complexing of radium with organic matter. Radium mobility and availability to plants could increase as a result. Schultz (1965) discusses the role of organic matter as a chelating agent for radium, thus alluding to the possibility that increased radium mobility may result.

Because of the potential to enhance radium mobility, further study on this question is recommended. If radium mobility were enhanced due to chelation with organic fractions, the threat to groundwater would increase.

6. Influence of Competing Ions on Radium Mobility

The presence of any ion in soil solution and on the exchange complex is dictated by the relative quantities of other similar ions in solution. This is especially true for ions of like charge, radius and activation energy. This is a dynamic situation because ions compete for exchange sites and seek equilibrium conditions while plants attempt to take up ions. It is known that excessive quantities of one ion may lead to nutritional imbalances and sometimes plant nutrient deficiencies. An example of this is the complimentary ion effect in plants where excess potassium acts to reduce the uptake of calcium and magnesium) (Tisdale, 1975).

The mobility of the radium ion, like any other, is apt to be affected by competing ions. Since it is present in extremely small quantities, it is expected to be proportionately more affected. This is especially true in the case of radium since it is similar to calcium and magnesium which are present in soils in much greater quantities. Schultz (1965) discusses such effects in relation to radium in a broad treatment of radionuclides. Complimentary ions will, due to like charge and valence, suppress the mobility of radium in soil solution thus plant uptake.

In this survey, no evidence was found to indicate that calcium limited radium uptake by plants. Radioactivity in plants was fairly similar even though a large amount of variation existed between sites with regard to soil calcium (Table 7). While no effect was attributed to calcium in this survey, it is likely that it is a controlling variable not detected in the limited number of observations. Experimental evidence has been accumulated in soils, substrates and in solution culture. Enough evidence is available to indicate that the regulating effect calcium exerts on radium plant uptake should be exploited. Thus, the effects of ionic competition and the tendency of radium to be strongly adsorbed by soil and sometimes discriminated against by plants in their uptake combine to decrease plant uptake of radium.

Although evident in the literature, the availability of a management opportunity due to competing ions was not indicated in this survey due to limited scope. Therefore, it is recommended that soil surveys be consulted and soils at proposal sites be analyzed for calcium and magnesium content. When possible, soils which possess high calcium and magnesium and/or possess calcium or magnesium carbonate layers should be favored in site selection.

7. Influence of Cation Exchange Capacity

Several studies have established that cation exchange capacity (CEC) is an important parameter in predicting the retention of applied metals and other ions (Logan and Chaney, 1983). Likewise, the CEC is cited as a factor in evaluating the soil's capacity to retain radium (Schultz, 1965) and has been generally acknowledged as a factor influencing radium retention (Marple, 1980; Osborne, 1965).

In this survey, a relationship between increasing cation exchange capacity and soil radium activity was observed (Table 8). Soils with CECs greater than 16 millequivalents per 100 grams (meq/100g) had radium activities more than twice as high as those with CECs less than or equal to 8 (meq/100g). This relationship must be viewed in concert with the increasing clay content. This is because increasing CEC may be related to increased clay content and clay may have a significant native radium content. But since CEC is employed to regulate the application of metal to sludge disposal sites, it should be useful in regulating radium loading to soils.

It seems apparent that cation exchange capacity offers a good management opportunity. It also provides the additional advantage of being a commonly and easily obtained parameter. Thus, its use as a management factor is recommended. Soils with a cation exchange capacity of less than nine should not be used for disposal of radium containing sludge. It is recommended that, whenever possible, sites used for sludge disposal should possess a cation exchange capacity of at least 15 meq/100 g.

8. Influence of pH

Soil reaction (pH) has long been used as a primary indicator to evaluate a range of soil capabilities. Nutrient availability and numerous crop growth factors are linked to soil pH. More recently, pH has become more important in evaluating a soil's waste treatment capability. Soil pH influences the retention and mobility of metals applied on land, and therefore their availability to plants.

The pH is a factor which affects a variety of soil properties and is employed to most consistently (CAST, 1980) evaluate a soils ability to protect food chain crops against metals -- especially cadmium -- by governing plant uptake. Likewise, the soil pH plays a major role in regulating plant uptake of radium by influencing the tenacity with which it is held by soils (Schultz, 1965) Kuneshava (1959) found that plant tissue obtained from acid soils had higher radium concentration than tissue obtained from more neutral soils. Soil pH affects radium mobility and, therefore, its potential threat to groundwater.

In the wastewater survey, site soils ranged from a pH of 5.8 to 7.2 (Table 9). As can be seen, no pH effect on plant uptake of radium is readily discernable. In all but one case of measurable radium activity in plants, the pH exceeded 6.5. However, it is interesting to note that the one exception (pH = 6.1) did have comparable radium activity in plant tissue to those obtained in soils with three times or more soil radium activity (Table 7). This indicates greater mobility of radium at a lower soil pH. To adequately define the relationship between pH and plant uptake of radium, a large sample population beyond the scope of this survey would have been necessary.

Currently, state regulations (NR 204) require that a sludge application site must have a pH of 6.5 in the soil/sludge mixture. This reflects the importance of pH in the management of heavy metals. Work aimed at improving acidic wastewater quality from uranium mill tailings demonstrated that the maintenance of a pH between 6.5 and 7.3 immobilized radium (Opitz, et al., 1985). Therefore, it is prudent to require that the soil pH at sites used for disposal of radium-containing sludge be maintained between 6.5 and 7.3.

9. Plant Uptake of Radium

a. Compartmentilization Within Plants

After radium is taken up by a plant it may stay in the root, be translocated and deposited in the shoots or leaves, or be transported and deposited in the fruit or grain. In general, roots have the greatest radium activity, followed by shoots and leaves. The least radium activity is found in the fruit or grain. The deposition of radium in plants is incompletely understood. A recent study summarizes the state-of-the-art in terms of radium dynamics in plants as follows: "The mobility of radium-226 within plant tissue appears to be high during transport from root to shoot but low after deposition within leaf tissues, resulting in an arcopedal concentration gradient. A potential for uptake by animals feeding on forage or leafy vegetables exists. Dynamics of translocation to nonleafy edible portions of plants has not been well documented." (Watson, 1983)

Roots consistently have higher radium activity than other plant parts when grown in both soil and substrate. This tendency was affirmed in a radium-spiked nutrient solution in which pea roots contained more than twice the radium activity of aerial parts (Kirchmann and Berino, 1965).

Data in this survey agree with the trend on partitioning, since fruit or grain were not found to have radium activity that could be separated from the background. No data for roots were obtained. However, four observations in three separate species showed radium activity in above ground plant components (Table 7). This activity was higher than the radium activity level in the "average plant" worldwide, but much less than agronomic plants in Poland or plants grown on uranium tailings (Table 2). Obviously, the ultimate use of the plant part in question must be considered. The use of root crops as of plants grown for consumption by dairy animals must be scrutinized.

b. Plant Age

It has been found that plant age affects its tissue radium activity. Grzybowska (1974) found that in agronomic plants radium activity decreased over the growing season. One study showed that when older plant leaves were compared to younger leaves, the younger leaves had a higher radium activity (Verkhovskaya, 1966). Plant samples in this survey were obtained only once (during autumn), so no local information on plant age has been generated through this survey.

c. Species Differences

No species differences were observed in this survey. Corn, soybeans and alfalfa had roughly the same radium activity. However, species differences are frequently reported in the literature. In one study, orchard grass had higher radium activity than alfalfa when grown on three separate soils (Grzybowska, 1974). When a grass and a shrub were grown on uranium tailings, the grass contained much higher radium activity than the shrub (Marple and Potter, 1982). Food crops such as cereal, potatoes, and leafy vegetables have been found to have measurable radium activity (Oakes and Shank, 1977).

d. Plant Radium Activity Related to Soil and Substrate

The radium content of substrate proved to be of importance when *Atriplex*, *Kochia*, and *Sitanion* were grown in uranium mill tailings. Twelve, 150 and 100 times the radium activity, respectively, were detected in plant tissue shown in tailings than when grown in local soils (Marple, 1980).

When plants were grown in soils with relatively high radium activity (3.9), alfalfa and orchard grass had radium activities of 3.1 and 4.8 pCi/gram, respectively. When grown in soils of 2.7 pCi/gram, plant tissue radium activities decreased to 2.7 and 4.4 pCi/gram, respectively, (Grzybowska, 1974).

In this survey, corn grown on soils with radium activities of 1.78 and 0.64 pCi/gram had radium activities of 0.023 and 0.018 pCi/gram, respectively (Table 7). No direct relationship to soil radium activities was apparent. In the observations of soybeans and alfalfa obtained, similar radium activities in soil and plant tissue radium activity were found. Thus, soils with radium activity levels varying from 0.51 pCi/gram to 1.57 pCi/gram yielded similar plant radium activity levels. Not enough data exists to establish a definite reason for this, but it is probably related to variations in soil factors such as pH, calcium content and organic matter (Table 7).

Radium activity in plants generally can be expected to increase with increasing substrate radium activity. Exceptions to this may be expected when variation in other soil factors is encountered.

D. Effect of Sludge Treatment and Mode of Application

This survey also addressed the question: could factors such as sludge application and sewage sludge digestion be linked to radium behavior as observed in the field? No relationship was observed between digestion or landspreading method and radium mobility.

E. Hazard Due to Surface Runoff

A greater hazard may exist due to erosion of soil/sludge mixtures containing radium than from its movement into groundwater. This is because radium is expected to adhere to soil particles (Guimond, 1978; Menzel, 1968; Schultz, 1965) and is susceptible to loss during surface erosion. It is recommended that disposal of sludge containing radium be limited to injection or surface application with incorporation. Also, disposal of sludge containing radium should be avoided under adverse climatic conditions.

F. Strategy for Control of Radium Mobility

Many soil factors affect plant uptake of isotopes, including radium. These include organic matter, pH, cation exchange capacity, the presence of competing ions, and clay content. Plant species differences are expected to occur. It appears, from limited data and from the literature, that the plant barrier impedes radium transfer in the food chain.

Radium is not concentrated in plants relative to the soil level (Menzel, 1965). Guimond (1978) indicates that radionuclides in plant tissue are excluded various degrees (relative concentrative factor does not usually exceed 0.1). Therefore, plant radium activity levels cannot exceed soil radium activity levels. This may allow for the formation of a control strategy. By setting conservative ceiling levels for soil radium, plant levels can be controlled. Also, by augmenting the maximum soil level approach with optmium selection of landspreading sites based on relevant soil variables, the threat to food chain crops can be further minimized. Obviously, the ultimate use of the plant material must be considered.

In cases where the entire above-ground plant is to be harvested or grazed, additional caution is necessary.

V. CONCLUSIONS

1. Sludge radium-226 activity was somewhat related to the radium activity in treatment plant influent and also to the sludge's capacity to adsorb and retain radium. Organic matter clearly displays a great capacity to adsorb and retain radium.

2. The radium-226 activity levels in the sludge in this survey were similar to common agricultural phosphate fertilizer and coal ash.

3. No hazard due to radon gas is expected on agricultural sites used for disposal of sewage sludge containing radium. This is because well ventilated conditions are expected. Risk is further minimized since the soil radium activity limits which have been established for sludge disposal sites are lower than the existing federal standard.

4. No elevation of soil radium activity could be attributed to landspreading of wastewater sludge containing radium. Soil radium differences are attributable to existing soil factors related to the kind and weathering of parent material, especially to clay content. Secondly, soil radium levels are influenced by the application of phosphate fertilizers containing radium. The influence exerted by fertilizers is highly variable and is a result of a wide range of fertilizer types, sources, and application rates.

5. Soil radium levels in both control sites and sludge disposal sites are similar to the worldwide average radium activity level. The soil radium levels detected in this survey were substantially less than levels found in western Europe.

6. It is anticipated that sludge application would cease due to current cumulative heavy metal loading limits before the soil radium activity is increased beyond what is expected from the application of phosphate fertilizer.

7. Radium mobility in soils is reduced to a degree dictated by the presence of competing ions. Further, it is likely to be held in soils more tightly than other alkaline earth cations.

8. Radium mobility is limited by the soil reaction. Radium is largely immobilized at a pH between 6.5 and 7.3.

9. Soil radium activity in this survey was strongly related to clay content. Clays have a great capacity to retain radium. Selection of disposal sites that have clay soil textures will exert a limiting effect on plant radium activity since plant uptake is influenced by radium mobility.

10. No evidence of downward migration of radium applied with wastewater sludge was detected. When radium activity in the lower increment exceeded that in the upper increment, it was a function of increased clay in the lower increment. Downward migration of radium is expected to be impeded by clay content, cation exchange capacity, maintenance of optimum pH range, organic matter, and the presence of competing ions. Threats to groundwater could arise if disposal sites with improper soil characteristics were employed, excessive rates of sludge containing radium were applied, or possibly if radium mobility was enhanced due to its being incorporated or chelated by the organic fraction.

11. Plant radium activity levels rarely exceeded background. When they did, radium activity in plant tissue was greatly below the soil activity level. Radium activity levels exceeding the background were not found in fruit or grain. Plants display an arcopetal concentration gradient for radium. Roots have the highest radium activity followed by shoots and leaves and lastly fruit or grain.

12. Plant radium activity was higher than the worldwide average for plants but much lower than that found in agronomic plants in Europe or in plants grown on uranium tailings.

13. No species differences in radium activity were observed. Based on the literature, it is expected that such differences will be detected with increased monitoring.

14. Plant radium activity was affected less by soil radium activity than by other soil factors which influence radium mobility.

15. The most likely threat to the environment as a result of the application of sludge containing radium in Wisconsin would be associated with soil erosion at sludge application sites and resulting in sedimentation.

16. In this survey, the method of landspreading or type of digestion of sludge had no apparent effect on radium.

VI. Recommendations

1. The disruption of landspreading of wastewater sludge containing radium is not warranted. Nevertheless, more stringent criteria should be employed in site selection. In addition, monitoring activity should increase in any landspreading program where the sludge radium-226 activity exceeds 2 pCi/gram.

2. The soil radium activity level at any disposal site should be limited to 2 pCi/gram. This level is to stay below the federal standard for adequate protection against radon gas. It will ensure that plant radium activity levels are maintained well below the soil ceiling level.

3. All efforts should be made to minimize the threat to receiving waters due to surface erosion of the soil/sludge mixture.

4. A strategy for controlling the entrance of radium into the food chain should be established and implemented. Since plants do not concentrate radium against the soil gradient, the maximum plant level can be controlled by establishing ceiling levels in soils. Many soil factors such as competing ions, pH, CEC, clay content and organic matter decrease radium mobility hence availability to plants. Therefore, through the control of soil radium levels and selected soil variables at the application site, acceptable plant radium activity levels can be maintained.

5. The Department should perform follow up studies addressing questions raised by the survey. Special emphasis should be placed on plant activity levels as controlled by soil factors. Leaching studies should be performed in columns, and the radium content of leachate should be determined under varying conditions. Finally, an evaluation of the potential that radium transport of groundwaters may be enhanced by virtue of its chelation by sludge or soil organic fractions should be made.

VII. GUIDELINES FOR THE APPLICATION OF RADIUM-226 CONTAINING WASTEWATER SLUDGE

It is recommended that the following set of guidelines be employed for the selection, operation, and monitoring of sites proposed or in use for the disposal of radium containing sludge. These guidelines will apply to sludges with a radium-226 content greater than 2 pCi/g. Radium-228 is not being considered at this time.

<u>Guidelines</u>

1. To be considered for radium sludge disposal, a proposed application site shall have a pH of between 6.5 and 7.3 prior to the application of sludge containing radium. Further, this pH shall be maintained during the period the site is used for the application of sludge containing radium.

2. To be considered for the disposal of sludge containing radium, a site must have soils with a cation exchange capacity of at least 10 meq/100 grams.

3. To be considered for the disposal of sludge containing radium, a soil must have a clay content of at least 25% in the upper 15 cm.

4. To be considered for the disposal of sludge containing radium, a soil must have an organic matter content of at least 12 tons per acre.

   Some flexibility in the site selection process is warranted since we are looking at the overall capability of the site and soil to afford adequate protection (retention). Therefore, we do not want to exclude an otherwise viable site based on its inability to meet one or more of the selection criteria. The following decision making criteria are available to provide adequate flexibility. This set of criteria <u>may</u> <u>not</u> be applied to soil pH.

   a. If a soil variable, which would exclude the site from selection, is 80% of the guideline value, it may still be approved if all the other selection criteria are satisfied.

      Example: If soil CEC is 8 meq/100 grams (80% of required 10 meq/100) but soil clay content is at least 25% in the upper 15 cm and soil organic matter satisfies the 12 tons per acre criterion it may be approved.

   b. In cases where two soil variables do not meet the selection criteria, they must be at least 90% of the guideline value for the site to be considered for approval. The site may still be approved at the discretion of the sludge manager. Written rationale must accompany such approvals.

5. Background soils data shall be obtained before site approval can be issued. Both the flow layer and the next underlying layer (approximately 15 cm) shall be sampled independently. Composite samples shall be obtained for each layer. About 1000 grams of soil should be collected. The number of composite samples obtained and the number of subsamples which constitute a sample are based on the total acreage applied for and the expected soil variability. These are given in the following table.

Field Size In Acres	Number of Separate Composite Samples Required	Number of Subsamples Necessary to Constitute Each Composite Sample
0-10	1	5
11-40	2	6
41-99	3	8
100 or more	3-5*	9

* Depends on soil variability

Analysis for pH, cation exchange capacity, organic matter, particle size analysis, phosphorus, potassium, calcium, magnesium, and radium-226 shall be performed and results shall be included in the application for site approval.

6. After every 3 years of site use for the disposal of sludge containing radium or after 5 years have elapsed from the initial sample date, the site shall again be sampled as in number 3 above. (In cases where the site is not used three times the applicator and generator shall notify the Department of termination of use and the site approval will be promptly revoked. In addition, the site shall undergo final sampling as in #3 and as described below.)

   Composite plant samples shall be obtained and analyzed for radium-226. The number of plant samples obtained shall be identical to the number of subsamples needed to constitute a composite soil sample and shall yield a wet weight of approximately 4 pounds. The entire above ground plant shall be obtained as near as possible to the soil subsample site. Plant samples shall then be segregated between above ground tissue (stems, stalks, petioles, leaves) and the "edible" portion (fruit, grain, or seed).

7. When data show that soil radium-226 activity has reached 2 pCi/g in any soil layer, sludge disposal on that field shall be terminated.

8. No site approved for disposal of sludge containing radium-226 shall be used for tobacco production.

9. No winter application of sludge containing radium shall be permitted. In cases where storage is inadequate, variances may be granted based on justification of need and shall be accompanied by a schedule for securing adequate storage capacity.

10. It is best if sludge is injected into soils. However, when it is necessary to surface spread sludge, it must be incorporated on the day it is landspread.

    General sampling guidelines and a list of laboratories which currently have the capability to perform the analysis are available.

Recommended Site Characteristics and Practices

1. Soil texture should be clay loam, silty clay loam, sandy clay, or silty clay material over bedrock or seasonal high groundwater. Should other textural zones be encountered, they should be evaluated on a case-by-case basis to insure that they provide an equivalent protection. This includes a demonstration that adequate available water holding capacity exists based on the depth to the limiting factor(s).

2. Disposal sites with slopes exceeding 6% should be avoided for disposal of sludge containing radium.

3. Disposal sites which are capable of affording maximum separation to seasonal groundwater and bedrock should be favored. At a minimum, a three foot separation between seasonal high groundwater and bedrock shall be provided. A five foot separation is favored.

4. Where possible, sites with high soil calcium levels shall be favored for the disposal of sludge containing radium.

5. Sites shall not be used for disposal of sludge containing radium when: the soil is saturated, standing water is observed, unfavorable weather is anticipated, the ground is frozen, or any condition is present which reduces the capability of the applicator to insure that sludge is uniformly injected or incorporated.

7145V

VIII. APPENDIX I

TABLE 1

Radium-226 in Influent, Effluent and Wastewater Sludge

Community	Influent pCi/l	Effluent pCi/l	Sludge pCi/g dry wt.	% Radium Retained in Sludge
Berlin	5.3	0.2	11.96	96.2
Mayville	<1.0	0.4	10.1 7.4	60
Menomonie	2.3	<1.0	24.1	>57
Mukwonago	3.8	0.4	32.78	89.5
Peshtigo	<1.0	<1.0	2.1	--
Union Grove	2.9	0.8	6.71	72.5

7145V

TABLE 2

Radium-226 (pCi/g dry weight) in Soils, Sludge and Other Common Materials

Materials	Range	Average	Source
Normal Superphosphate Fertilizer from Florida		21.0	Guimond (1978)
Ammonium Phosphate Fertilizer		5	Guimond (1978)
Uranium Mill Tailings		415.0	Marple (1980)
Wisconsin Wastewater Sludge	2.1-36.1	17.43	Survey results
Worldwide Soils	0-17,000	0.8	Brooks (1972)
United States Soils	0.1-3.0	--	Guimond (1978)
Wisconsin Soils	0.29-1.36	0.81	Survey results
Wisconsin Sludge Site Soils	0.29-1.78	0.91	Survey results
Soils in Great Britian	0.5-2.1	1.3	Vassilaki et al. (1966)*
Soils in Italy		0.72	Bortoli & Gaglione (1972)
Soils in Poland		3.1	Grzybowska (1974)
Calculated Maximum Permissible Soil Level Based on Exposure		220	Hollins (1977)
Average plant (worldwide)		0.004	Osburn (1965)
Corn in Poland		3.1	Grzybowska (1974)
Alfalfa in Poland		4.8	Grzybowska (1974)
Soybean plant grown on sludge field in Wisconsin		0.027	Survey results
Plant Grown on Uranium Tailings		50.0	Marple & Potter (1981)
Coal Ash		24.0	Eisenbud & Petrow (1964)
Milk from Wisconsin		0.24*	Watson et al. (1984)
Petroleum		0.49	Eisenbud & Petrow (1964)

* Value is in pCi/kg

TABLE 3

Radium-226 (pCi/g dry weight) Levels in Soils at Sludge Sites and Corresponding Control Sites

Community	Sludge Site	Control Site	Sludge Site Exceeds Control Site
Berlin	0.51	0.29	*
	0.32	0.31	*
	0.29	0.29	
	0.38	0.31	*
Mayville	1.08	1.14	
	0.98	1.11	
	0.92	0.85	*
	0.99	0.71	*
Menomonie	1.00	0.48	*
	0.86	0.42	*
	0.41	0.42	
	0.34	0.44	
Mukwonago	0.64	0.91	
	0.58	0.95	
Peshtigo	0.57	0.58	
	0.55	0.65	
	0.48	0.52	
	0.54	0.49	
Union Grove	1.78	1.21	*
	1.49	1.36	*
	1.57	1.21	*
	1.57	1.36	*
	1.48	1.21	*
	1.50	1.36	*

7145V

TABLE 4

Radium-226 (pCi/g dry weight) in Upper Soil Increment Compared to the Lower Soil Increment

				Cases Where B exceeds A	
Community	Increment	Sludge Site	Control	Sludge Site	Control
Berlin	A	0.51	0.29		*
	B	0.32	0.31		*
	A	0.29	0.29	*	
	B	0.38	0.31	*	
Mayville	A	1.08	1.14		
	B	0.98	1.11		
	A	0.92	0.85	*	
	B	0.99	0.71	*	
Menomonie	A	1.00	0.48		
	B	0.86	0.42		
	A	0.42	0.41	*	
	B	0.44	0.34	*	
Mukwonago	A	0.64	0.91		*
	B	0.58	0.98		*
Peshtigo	A	0.57	0.58		*
	B	0.55	0.65		*
	A	0.48	0.52	*	
	B	0.54	0.49	*	
Union Grove	A	1.78	1.21		*
	B	1.49	1.36		*
	A	1.57	1.21		*
	B	1.57	1.36		*
	A	1.48	1.21	*	
	B	1.50	1.36	*	

TABLE 5

Influence of Soil Texture on Radium-226 Activity in Soil (Both Control and Sludge Sites)

Activity Ranges in Soil (pCi/g dry weight)	Observations by Textural Class											
	sa	losa	salo	lo	silo	si	sacllo	cllo	sicllo	sacl	sicl	cl
0.29-1.08	4	4	10	6	4	0	0	2	0	0	0	0
1.11-1.78	0	0	0	0	0	0	0	0	7	0	2	0

increasingly finer soil texture

7145V

TABLE 6

Influence of Organic Matter (tons per acre) on Radium-226 Activity (pCi/g dry weight) in Soil (Both Control and Sludge Sites)

Organic Matter Range	Average Ra-266 Activity
0-11	0.37
12-45	0.89
>45	1.27

7145V

TABLE 8

Influence of Soil Cation Exchange Capacity (CEC)
Millieqivalents per 100 g on Soil Radium-226 Activity
(pCi/g dry weight) (Both Control and Sludge Sites)

Range of CEC	Average Radium-226 Activity in Soil
< 9	0.45
9-15	0.90
> 15	1.07

7145V

TABLE 9

Influence of Soil pH on Ra-226 (pCi/g dry weight)
Activity in Soils (Both Control and Sludge Sites)

Soil pH Range	Average Soil Radium-226 Activity
5.8-6.2	0.91
6.3-6.6	0.66
6.7-7.1	0.92
> 7.1	0.78

7145V

TABLE 7

Plant Radium-226 Activity Levels Related to Selected Soil Variables

Community	Plant Species	Radium-226*** Activity in Plant (pCi/g)	Radium-226 Activity in Soil (pCi/g)	Soil pH	Soil Calcium (lbs/ac)	Soil CEC	Soil Clay %	Organic Matter
Berlin	Alfalfa	0.023	0.51	6.1	1300M	4	5	16
Mayville	*							
Menomonie	**							
Mukwonago	Corn	0.018	0.64	7.2	4450H	16	7	50
Peshtigo	**							
Union Grove	Corn	0.023	1.78	6.6	4800H	17	36	50
	Soybeans	0.027	1.57	7.0	1300M	8	31	45

* Radium-226 activity in plant tissue at or less than background

** No plant sample obtained

*** Value is for stalks, stems, leaves only; no detectable radium in grain or fruit

7145V

APPENDIX II

METHODS

Prescribed Procedures for Measurement of Radioactivity in Drinking Water
Method 903.1 EPA
Radium-226 in Drinking Water Radon Emanation Technique.

The radium in a one liter water sample is separated and concentrated by coprecipitation on lead and barium sulfate. The precipitate is dissolved in EDTA reagent, then placed in de-emanation bubbler which is sealed and stored to allow for the ingrowth of radon-222. After ingrowth (at least four days), the gas is purged into a scintillation cell which is counted for alpha activity. Calculations are done and results reported in pCi per liter of sample.

Tennessee Valley Authority Radiological Procedures Manual
Procedures RA-01
Radiochemical Determination of Radium-226 in Environmental Samples.

A four gram sample of ashed vegetation is fused with sodium hydroxide and sodium carbonate. The fusion cake is dissolved in boiling distilled water, and the resultant precipitate is dissolved in dilute hydrochloric acid. Twenty-five milligrams of resulting solution is then placed in a de-emanation bubbler, which is sealed and stored to allow for the ingrowth of radon-222. After ingrowth (at least 14 days), the gas is purged into a scintillation cell which is counted for alpha activity. Calculations are done and results reported in pCi per gram of dried vegetation.

Tennessee Valley Authority Radiological Procedures Manual
Procedure U-02
Uranium and Radium by Germanium Spectroscopy

A sample of dried and ground soil is placed in a tared 500 ml Marinelli beaker. If there is insufficient sample, salt (NaCl) is homogeneously added to bring the sample to volume. The beaker is weighted, then sealed and stored for at least 28 days to allow for the ingrowth of radon-222 daughter products. After ingrowth the sample is counted on a germanium-lithium (GeLi) gamma ray detection system for a minimum of two hours. The areas of the 295-keV and 352-KeV peaks of lead-214, and the 609 KeV peak of bismuth-214 are use in the calculations to determine the radium-226 activity in pCi per gram.

7145V

IX. BIBLIOGRAPHY

D. C. Adriano, K. W. McLeod, and T. G. Ciravolo. 1981. Plutonium, Curium and Other Radionuclide Uptake by the Rice Plant from a Naturally Weathered, Contaminated Soil. Soil Sci. 132(1): 83-88.

B. C. F. Bacs III, R. D. Sharp, A. L. Sjoreen, and R. W. Sbor. A Review and Analysis of Parameters for Assessing Transport of Environmentally Released Radionuclides Through Agriculture. USDOE Report ORNL-5786. Oak Ridge National Laboratory, in press.

M. De Bortoli and P. Gaglione. 1972. Radium-226 in Environmental Materials and Foods, Health Phys., 22(1):43-48 (January).

R. R. Brooks. 1972. Geobotany and Biogeochemistry in Mineral Exploration. New York. Harper and Row.

C. C. Delwiche, R. O. Hansen and P. R. Stout. 1958. Proceedings of the 2nd International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958. Vol. 18, p. 551 United Nations, New York. (Cited in Osburn, 1965).

CAST. 1980. Effects of Sewage Sludge on the Cadmium and zinc content of crops. Council for Agric. Sci. Tech. No. 83. Ames, IA. 77 pp.

R. B. Corey, 1985 Personal Communication. University of Wisconsin. Madison, Wisconsin.

C. C. Delwiche, R. O. Hansen, P. R. Stout, G. B. Jones and G. Ranzani. 1958. Proceedings of the 2nd International Conference on the Peaceful Uses of Atomic Energy, Geneva, Vol. 18,

M. Eisenbud and H. G. Petrov. 1964. Radioactivity in the Atmospheric Effluents of Power Plants that Use Fossil Fuel. Science 144:288-289.

Environmental Pipeline. 1986. Radon - A Real Radiation Problem. Vol. 4, No. 2. Tennessee Dept. of Health and Env. Nashville, Tennessee.

Federal Register, Vol. 48, No. 3, Wednesday, January 5, 1983. Rules and Regulations, Part 192. Environmental Protection Standards For Uranium Mill Tailings.

J. F. Gillott. 1979. Clays, Physical Chemistry in the Encyclopedia of Soil Science. Part 1. Edited by R. W. Fairbridge and C. W. Finkl, Jr. Dowden, Hutchinson and Ross, Inc. Stoudsburg, Penn.

D. Grzybowska. 1974. Uptake of ^{226}Ra by Plants from Contaminated Soils, Nucleonika, 19(1):71-78.

R. J. Guimond, 1978. The Radiological Aspects of Fertilizer Utilization in "Radioactivity in Consumes Prod". (1978) NUREG/CP-0001, U.S. Nuc. Reg. Comm. A. A. Moghissi, P. Paras, M. W. Curton, R. F. Barkes (ed.), August 1978.

N. A. Hallden and J. H. Harley, 1974. Radium-226 in Diet and Human Bone from San Juan, Puerto Rico, Nature, 204(4955): 240-241.

N. Hahn. Radium in Wisconsin Groundwater and Removal Methods for Community Water Systems; Wisconsin Department of Natural Resources; December, 1984.

R. H. Hesslein and E. Salavich. 1984. Geotechnical Pathways and Biological Uptake of Radium in Small Canadian Shield Lakes. Can. J. Fish and Aquat. Sci., Vol. 41, pp. 459-468.

H. G. Hollins, 1977. Radium in Vegetable Gardens. In: the Procedures of IV International Congress of International Radiation Protection Association, ed. G. Bresson, Vol. 3, pp. 1045-48. Montrevil:Imprimetic Gauthier-Vallaus.

R. B. Holtzman, P. W. Urnezis, A. Padova, and C. M. Bobula III, 1979 Contamination of the Human Food Chain by Uranium Mill Tailings Piles, NUREG/CR-0758 (ANL/ES-69). Argonne National Laboratory. Report 4.

T. J. Logan and R. L. Chaney. 1983. Utilization of Municipal Wastewater Sludge on Land-Metals. in Proceedings of the 1983 workshop on Utilization of Municipal Wastewater and Sludge on Land.

R. Kirchmann, A. LaFontaine, J. Van den Hock, and G. Koch, Comparison of the Rate of Transfer to Cow Milk of ^{226}Ra from Drinking Water and ^{226}Ra Incorporated in May. C. R. Sciences Soc. Biol. Ses. Fil 166(11): 1557-1562 (1972).

R. Kirchmann, and G. Berino. 1965. Compartmentalization of Ra-222 in *Pisum sativum*. Phyton 22:137-39.

K. G. Kunasheva. 1959. Trav. Lab. Biogeochem USSR. 16, 30.

E. A. Liegel, C. R. Simson, and E. E. Schulte. 1980. Sampling Soils for Testing. Cooperative Extension Programs, University of Wisconsin Extension. A2100.

T. J. Logan, and R. L. Chaney. 1983. Utilization of Municipal Wastewater and Sludge on Land - Metals in: Proceedings of the 1983 Utilization of Municipal Wastewater on Land. pp. 235-323. University of California, Riverside, CA.

M. L. Marple. 1980. Radium-226 in Vegetation and Substrates at Inactive Uranium Mill Sites, Report LA-8183-T, Los Alamos Scientific Laboratory, NTIS, January 1980.

Marple, M. L., and D. Potter, 1982. Uptake of radium-226 by plants of inactive uranium mill sites in the southwestern USA. In: environmental management of long-lived radionuclides, Int. Al Energy Agency IAEA. SM-257: 237-246.

E. Mazor. 1963. Israel J. Earth Science 12 & 41 (Cited in Menzel, 1968).

R. G. Menzel. 1968. "Uranium, Radium, and Thorium Content in Phosphate Rocks and Their Possible Radiation Hazard," J. Agr. Food Chem. 16, 231-234.

K. B. Mistry. 1963. Agricultural Research Council Radiobiological Laboratory, Report ARCRL. 10. HMSO, London.

E. L. Mordberg, V. M. Aleksandruk, G. F. Kovygin, I. I. Shevchenko, V. M. Blyumsbtein, and G. F. Yushkevick. 1976. Translocation of Isotopes of the Uranium-Radium Series into the Grain of Some Agricultural Crops, Chem. Abstr., 84: 134563 (1976), abstracted from Gig. Sanit. 2: 58-61.

J. L. Mortenson. 1963. Complexing of Metals by Soil Organic Matter. Soil Sci. Soc. Amer. Proc. 27:179-186.

H. Muth, B. Rajewsky, H. J. Hantke, and K. Aurand. 1960. The Normal Radium Content and the ^{226}Ra/Ca Ratio of Various Foods, Drinking Water and Different Organs and Tissues of the Human Body, Health Phys., 2: 239-245.

National Council on Radiation Protection and Measurements (NCRP). 1975. Natural Background Radiation in the United States, NCRP Report No. 45, 44-47.

Y. C. Ng. 1982. A Review of Transfer Factors for Assessing the Dose from Radionuclides in Agricultural Products, Nucl. Saf., 23(1): 57-71 (January-February).

T. W. Oakes, K. E. Shank, C. E. Easterly, and L. R. Quintana. 1977. Concentrations of Radionuclides and Selected Stable Elements in Fruits and Vegetables in Proceedings of the University of Missouri Annual Conference on Trace Substances in Environmental Health XI, D. D. Hemphill (Ed.), pp. 123-132, University of Missouri, Columbia.

B. E. Opitz, M. E. Dodson, R. Serne. 1985. Uranium Mill Tailings Neutralization: Containment, Complexation and Tailings Leaching Studies. Office of Nuclear Regulatory Research U.S. Nuclear Regulatory Commission. NRC FIN B2370.

M. E. Opitz, M. E. Dodson, R. J. Serne. 1985. Uranium Mill Tailings Neutralization: Contaminant Complexation and Tailings Leaching Studies. NUREG/CR-3906 PLN-5179, Pacif NW Labs. Battelle Mem. Inst. Prep to Nuc. Reg. Comm.

W. S. Osburn. 1965. Primordial Radionuclides: Their Distribution, Movement, and Possible Effect Within Terrestrial Ecosystems. Health Phys., 11:1275-1295.

G. V. Rusanova. 1964. Behavior of Radium and Calcium in the Soil Plant System. Soviet Soil Sci., 3:275-280.

D. R. Raymo, M. H. Momeni and C. Sabau. 1980. Forage Uptake of Uranium Series Radionuclides in the Vicinity of the Anaconda Uranium Mill, in Uranium Mill Tailings Management, Proceedings of the Third Symposium, Nov. 24-25, 1980, Colorado State University, Fort Collins, Co., pp. 67-66, Civil Engineering Department, Colorado State University.

P. D. Shandley. 1953. The Radium Content of Common Foods, Report UR-225, University of Rochester.

R. K. Schultz. 1965. Soil Chemistry of Radionuclides, Health Phys., 1317-1324.

O. Talibudeen. 1964. Natural Radioactivity in Soils. Soil Fert. 27:347-359.

A. I. Taskaev, V. A. Ovchenkov, R. M. Aleksakhim, and I. I. Shuktomova. 1977. Uptake of Radium-226 by Plants and Changes in its State in the Soil Above Ground Mass Litter-fall System, Pochvovedenie, 2:42-48.

S. L. Tisdale and W. L. Nelson. 1975. Soil Fertility and Fertilizers. Third Edition MacMillan Publishing Co., New York.

D. Telefair, R. Garrison and O. C. Smith. 1960. Science 131, 727.

M. Tsekos and D. M. Keller. 1983. Adsorption of Radium-226 by Biological Absorbents: Biotechnology and Bioengineering, Volume 25, pp. 201-215, Wiley and Sons.

M. Vassilaki, L. Salmon, and J. A. B. Gibson. 1966. Measurement of Radioactivity in Soil. Geochim. Cosmochim. Acta 30:601-606. (Cited in Marple, 1980).

P. P. Vavilov, O. N. Popova, and R. P. Kodaneva. 1964. The Behavior of Radium in Plants. Abstracted from Dokl. Akad. Neuk. SSSR 157:992-994. (Cited in Marple, 1980).

I. N. Verkhovskaya, B. I. Gruzdev, V. I. Maslov, K. I. Maslova, V. S. Nikiforon, V. J. Ovchenkov, O. N. Popova, D. M. Rubtsov and P. P. Vavilov. 1969. The Content and Translocation of Natural Radioactive Elements in the System "Soil-Plants-Animals" Under Natural and Experimental Conditions. In Actes du Symposium International de Radioecologic, Vol. 2, pp. 781-832. Cadarache: Centre d'Etudes Nucleaires. (Cited by Marple, 1980).

A. P. Watson, E. L. Etnier, and L. M. McDowell-Boyer. 1983. Radium-226 in Drinking Water and Terrestrial Food Chains: A Review of Parameters and An Estimate of Potential Exposure and Dose, USDOE Report on ORNL/TM-8597, Oak Ridge National Laboratory, NTIS, April.

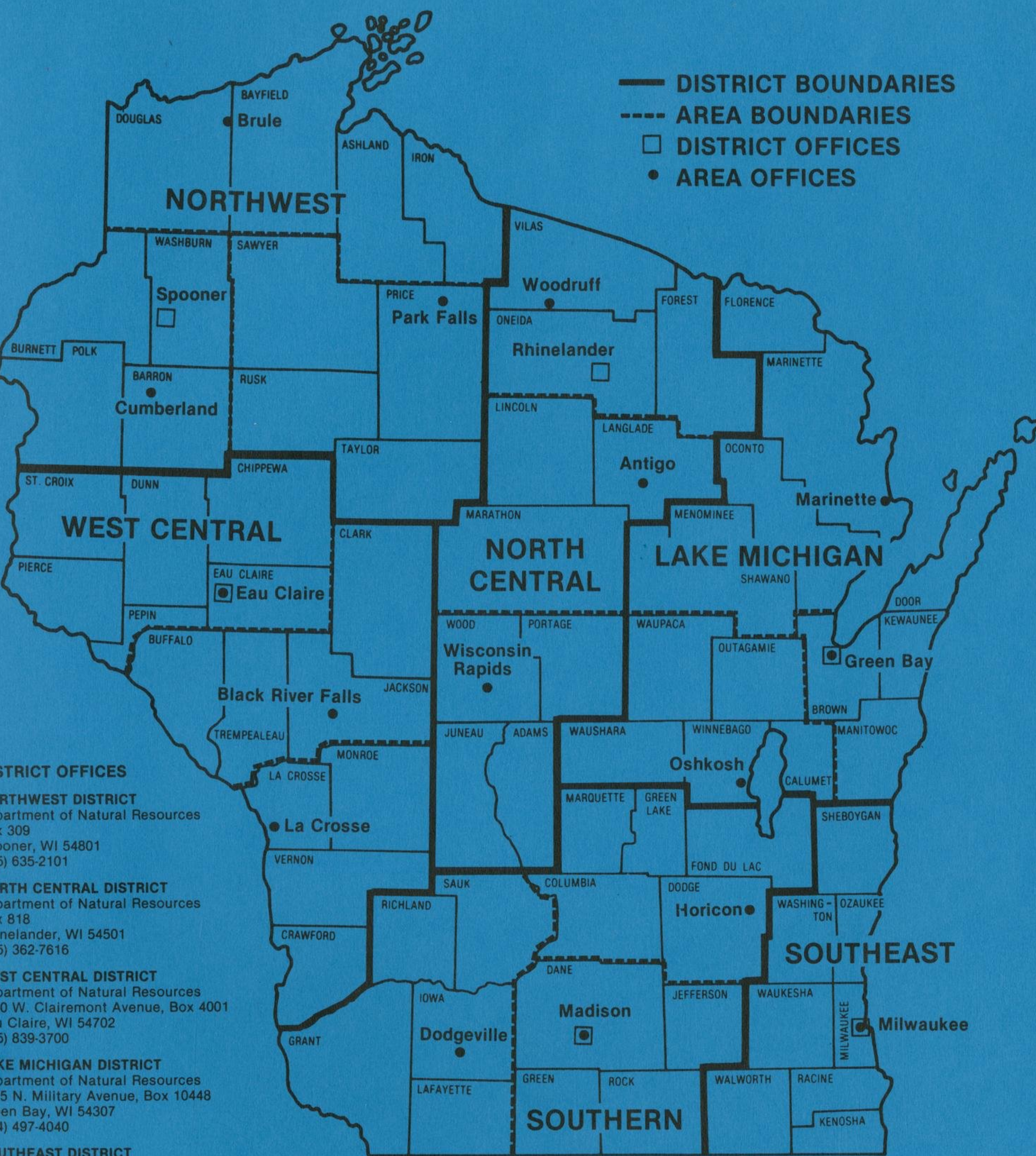
A. R. Williams. 1982. Biological Uptake and Transfer of Radium-226: A Review in Environmental Migration of Long-Lived Radionuclides. Int. At. Energy Agency IAEA-SM 257:207-222.

M. Williams. 1985. The Fate of Radium-226 and Radium-228 in the Wastewater Treatment Process. A survey. Wisconsin Dept. of Natural Resources, Bureau of Solid Waste Management.

A. P. Watson. 1984. Ra^{226} in Drinking Water and Terrestrial Food Chains: Transfer Parameters and Normal Exposure and Dose. Nuclear Safety Vol. 25, No. 6.

7145V

DNR FIELD DISTRICTS AND AREAS



DISTRICT OFFICES

NORTHWEST DISTRICT
Department of Natural Resources
Box 309
Spooner, WI 54801
(715) 635-2101

NORTH CENTRAL DISTRICT
Department of Natural Resources
Box 818
Rhinelander, WI 54501
(715) 362-7616

WEST CENTRAL DISTRICT
Department of Natural Resources
1300 W. Clairemont Avenue, Box 4001
Eau Claire, WI 54702
(715) 839-3700

LAKE MICHIGAN DISTRICT
Department of Natural Resources
1125 N. Military Avenue, Box 10448
Green Bay, WI 54307
(414) 497-4040

SOUTHEAST DISTRICT
Department of Natural Resources
2300 N. Dr. Martin Luther King, Jr. Drive
Box 12436
Milwaukee, WI 53212
(414) 562-9500

SOUTHERN DISTRICT
Department of Natural Resources
3911 Fish Hatchery Road
Fitchburg, WI 53711
(608) 275-3266

Rev. 8-85

OUR MISSION:

To protect and enhance our Natural Resources —
our air, land and water;
our wildlife, fish and forests.

To provide a clean environment
and a full range of outdoor opportunities.

To insure the right of all Wisconsin citizens
to use and enjoy these resources in
their work and leisure.

And in cooperation with all our citizens
to consider the future
and those who will follow us.

Wisconsin
Dept. of Natural Resources

050854- Fate and Mobility of Radium-226 in Municipal Wastewater Sludge Following Agricultural Landspreading: A Survey