United States
Environmental Protection
Agency

Office of Water
Regulations and Standards
Washington, D.C. 20460

Water

April, 1986

EPA

Radioactivity of Municipal Sludge

RADIOACTIVITY OF MUNICIPAL SLUDGE

U.S. Environmental Protection Agency
Office of Water Regulations and Standards
Wastewater Solids Criteria Branch
April, 1986

PREFACE

Section 405 of the Clean Water Act requires the U.S. Environmental Protection Agency to develop and issue regulations which: (1) identify uses for sludge including disposal; (2) specify factors to be taken into account in determining the measures and practices applicable to each use or disposal (including costs); and (3) identify concentrations of pollutants which interfere with each use or disposal. In order to comply with this statutory mandate, EPA has embarked on a major program to develop four major technical regulations: land application including distribution and marketing, landfilling, incineration, and ocean dumping. EPA has also developed proposed regulations which govern the establishment of state sludge management programs to implement both existing and future criteria.

During the regulatory development process, questions related to radioactivity in sewage sludge were presented:

(a) What radioactive compounds are present in sewage sludge and at what levels;

(b) Is there the potential for health problems from the reuse and disposal of such sludges; and

(c) Are there ways of limiting radioactivity in sewage sludges.

The purpose of this document is to summarize the available data on the radioactive constituents and levels found in sewage sludges. This compilation of data may be used in the future to evaluate any potential health concerns once risk assessment methodologies for radioactive materials in sludges are developed. EPA would appreciate any comments and additional data on radioactive compounds in sewage sludge. Any questions related to this document may be directed to:

Elliot Lomnitz
U.S. Environmental Protection Agency
Wastewater Solids Criteria Branch (WH-585)
Washington, DC 20460

Patrick M. Tobin

Patrick M. Tobin, Director
Criteria and Standards Division
Office of Water Regulations
and Standards

TABLE OF CONTENTS

INTRODUCTION 1

APPROACH 1

RESULTS 2

SUMMARY 21

REFERENCES 23

APPENDIX A A-1

APPENDIX B B-1

APPENDIX C C-1

APPENDIX D D-1

INTRODUCTION

Section 405 of the Clean Water Act requires EPA to issue and publish regulations related to the reuse and disposal of municipal sludge. The Criteria and Standards Division is responsible for publishing regulations on four major reuse/disposal options: land application including distribution and marketing, landfilling, incineration, and ocean dumping. During the course of the regulatory development process, the issue of radioactivity in sludges has been raised as being sufficiently important to warrant investigation. The reasons for concern were the public or occupational health risks that could potentially result from the various disposal options if radionuclides were found at high concentrations in sludge. For example, landspreading of contaminated sludges on agricultural crops could result in exposure to humans through bioaccumulation of radionuclides in the foodchain.

The purpose of this study was to determine the extent of existing data on levels of radioactivity in municipal wastewater treatment sludge. Assessment of the available data will be conducted by the U.S. EPA in the future to determine if radionuclides in sludge constitute a problem. This assessment would commence once risk assessment methodologies for radioactivity in municipal sludge are developed.

APPROACH

Two methods were used to identify and obtain data on radionuclides in municipal wastewater treatment sludges. The first consisted of searching computerized literature data bases for pertinent articles. The data bases searched included Agricola, Aqualine, Aquatic Science Abstracts, Biosis Previews, Chemical Abstracts, DOE Energy Data Base, Enviroline, NTIS, Pollution Abstracts, and Water Resources Abstracts. The years covered by each data base are given in Appendix A. In most cases the data bases were searched as far back as the data base extended, usually the late 1960s or early 1970s.

The search strategy consisted of matching the keywords "sludges" and "wastewater treatment" against "radioactive", "radionuclide", "beta", "gamma", "ionizing", and "radiation". The search was further narrowed by limiting with the keywords "municipal" and "city". In some data bases, numerous articles appeared in the searches which examined the use of gamma irradiation to sterilize sewage sludge. In these cases, a further screening was performed to eliminate studies keyed as "irradiation".

The computer searches identified several studies containing data on radioactivity associated with municipal wastewater treatment sludges. Pertinent articles were obtained from local libraries, where possible, and through interlibrary loan. Upon receipt, the articles were reviewed further for any pertinent references cited in them.

The second method used to obtain data was a telephone survey. The purpose of the telephone survey was to identify additional data not identified in the computer data base searches, particularly those data which were not available in the published literature. To conduct the telephone survey, a form containing a standard set of questions was developed. This form appears in Appendix B. Interviewees were identified by contacting agencies known to regulate municipal wastewater or radionuclides and by requesting recommendations from other interviewees. Approximately 90 individuals were interviewed in the telephone survey. A list of interviewees is also provided in Appendix B. These interviewees represented U.S. EPA regional offices, the U.S. Nuclear Regulatory Commission, U.S. Department of Energy, national laboratories, state environmental and health protection agencies, municipalities, and sludge distributors. The telephone survey was successful in identifying additional information on radioactivity in sludge, including both published and unpublished data.

The staff involved in data collection and preparation of this document are listed in Appendix D.

RESULTS

The potential sources of radioactivity in sludges are shown in Figure 1. Studies containing data on radioactivity levels in municipal wastewater sludges are summarized in Table 1. The studies ranged from those reporting data for numerous radionuclides to those examining specific radionuclides from specific sources. The studies presented in Table 1 were

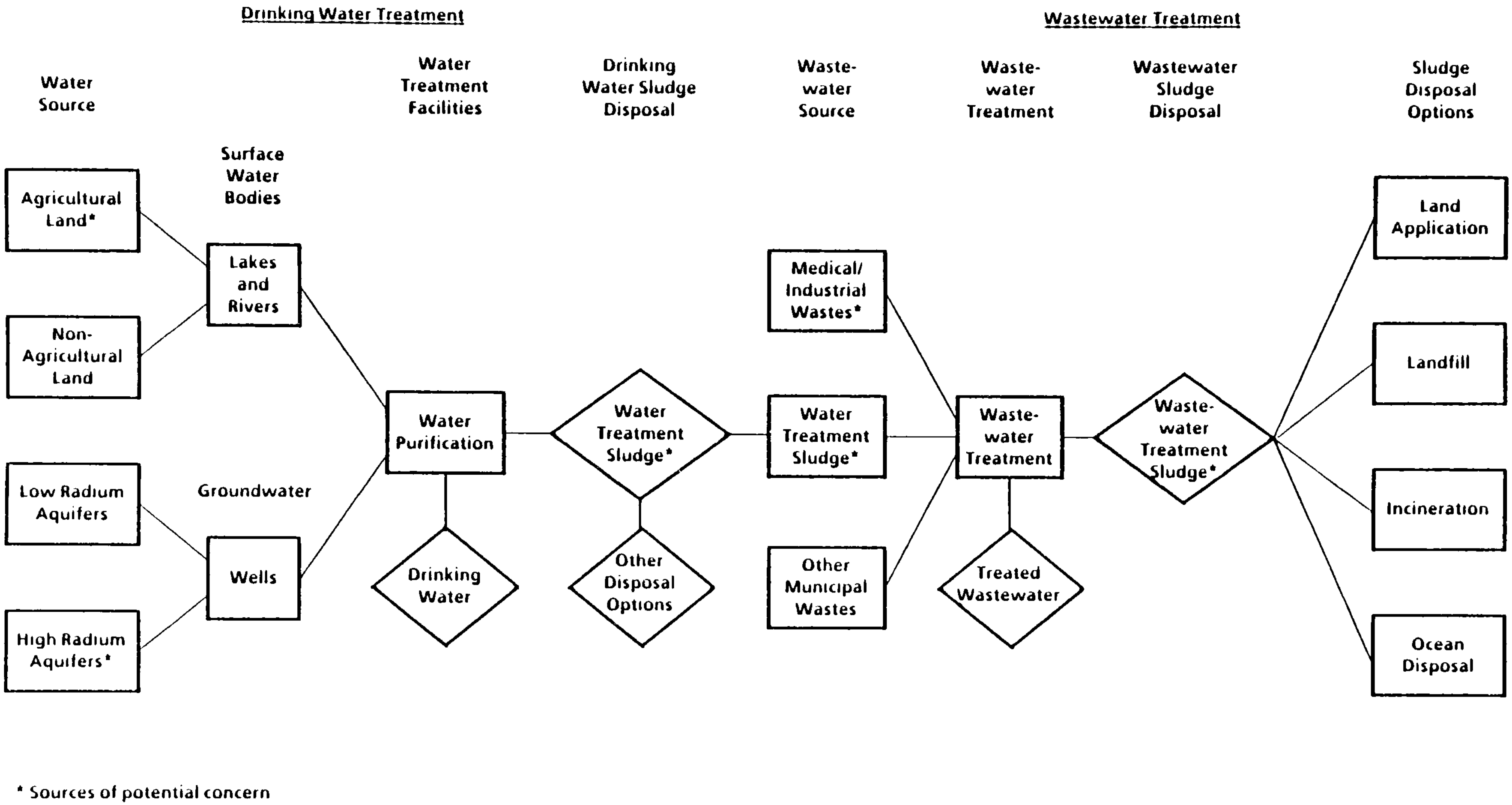

FIGURE 1. GENERALIZED DIAGRAM OF RADIOACTIVITY SOURCES ENDING IN MUNICIPAL WASTEWATER TREATMENT SLUDGES

TABLE 1. SUMMARY OF RADIOACTIVITY LEVELS IN MUNICIPAL WASTEWATER TREATMENT SLUDGES

Study	Location	Type of Sludge	Sample Period	Radio-nuclide	$T_{1/2}$	Radioactivity	Analytical Technique	Sources of Contamination	Comments/ Conclusions
Prichard et al., 1981	Sims Bayou Sewage Treatment Facility, Houston, Texas	Conventional activated sludge--flash dried	88 days--Spring to Summer, 1975	^{131}I	8d	∿7 pCi/g to ∿180 pCi/g (most data points < 50 pCi/g, estimated from graph)	Gamma spectroscopy, Covell's method for net count under photopeak	9 institutions with nuclear medicine programs served by sewage plant	^{131}I dominated gamma spectra for dried sludge and effluent. Ratio of ^{131}I in sludge to ^{131}I in effluent was 1:3.3. Study focused on estimating daily discharge (Ci/day) rather than conc. (Ci/g). Concluded no significant dose to population from sludge used as soil conditioner due to short $T_{1/2}$ of ^{131}I.
		Total discharge from treatment plant (sludge + effluent)		^{131}I ^{99m}Tc	8d 6h	4.4 mCi/day 2.6-11.8 mCi/day $\bar{x}$ = 6.9 mCi/day			
Alberts and Wahlgren, 1977	Chicago Metropolitan Sanitary District, Stickney, Illinois	Sediment from Imhoff Process, air-dried and aged in large open-air piles, distributed to public for free as "Nu Earth" fertilizer	Summer, 1974	$^{239,240}Pu$ ^{241}Am	* 432y	28.5 fCi/g ash (21.4 fCi/g dry wt) 4.4 fCi/g ash (3.3 fCi/g dry wt) $^{239,240}Pu/^{241}Am$ = 6.5	Samples dried then ashed (75% ash/dry wt) then analyzed for $^{239,240}Pu$ and ^{241}Am	None specified	Ratio of $^{239,240}Pu/^{241}Am$ more representative of soil than lake sediment. Concentrations below background levels. Concluded there is little evidence that $^{239,240}Pu$ or ^{241}Am from sludge pose a health hazard.
			Spring, 1975	$^{239,240}Pu$ ^{241}Am	* 432y	17.0 fCi/g ash (12.8 fCi/g dry wt) 2.0 fCi/g ash (1.5 fCi/g dry wt) $^{239,240}Pu/^{241}Am$ = 8.5			
Alberts et al., 1974	Chicago Metropolitan Sanitary District, Stickney, Illinois	Sediment from Imhoff Process, air-dried and aged in large open-air piles, distributed to public for free as "Nu Earth" fertilizer	November, 1974	$^{239,240}Pu$ ^{137}Cs	* 30y	28.5 fCi/g ash (21.5 fCi/g dry wt) 1.43 pCi/g ash (1.08 pCi/g dry wt) $^{137}Cs/^{239,240}Pu$ = 50.0	Samples dried then ashed (75% ash/dry wt) then analyzed for $^{239,240}Pu$ and ^{137}Cs	None specified	Concluded that although significant amounts of radioisotopes may be conc. in sludge, material appears to be in form not readily available for removal and is unlikely to remobilize.

TABLE 1. SUMMARY OF RADIOACTIVITY LEVELS IN MUNICIPAL WASTEWATER TREATMENT SLUDGES (Continued)

Study	Location	Type of Sludge	Sample Period	Radio-nuclide	$T_{1/2}$	Radioactivity	Analytical Technique	Sources of Contamination	Comments/ Conclusions
The Metropolitan Sanitary District of Greater Chicago, 1979	Chicago - Lawndale Lagoons and Harlem Avenue disposal site	Lagoon sludge - "dried (non-volatile) material"	1978	Total alpha	NA	Average = 7.46 pCi/g dry wt n = 47 (Range 3.89 - 10.67 pCi/g dry wt)	Beckman Widebeta II proportional counter	None specified	Expected fluctuations observed; no surge of radioactivity in any one sample.
				Total beta	NA	Average = 54.5 pCi/g dry wt n = 47 (Range 39.9 - 102.6 pCi/g dry wt)			
			1979	Total alpha	NA	Average = 10.00 pCi/g dry wt n = 10 (Range 6.24 - 13.12 pCi/g dry wt)			
				Total beta	NA	Average = 78.9 pCi/g dry wt n = 10 (Range 45.7 - 102.5 pCi/g dry wt)			
The Metropolitan Sanitary District of Greater Chicago, 1982	Chicago - Lawndale Lagoons and Harlem Avenue disposal site	Lagoon sludge - "dried (non-volatile) material"	1980	Total alpha	NA	Average = 9.82 pCi/g dry wt n = 25 (Range 7.43 - 13.41 pCi/g dry wt)	Canberra alpha/ beta proportional counter	None specified	Fluctuations in radioactivity conc. observed, no surge of radioactivity in any one sample.
				Total beta	NA	Average = 59.6 pCi/g dry wt n = 39 (Range 48.3 - 126.0 pCi/g dry wt)			
			1981	Total alpha	NA	Average = 29.30 pCi/g dry wt n = 39 (Range 16.19 - 39.12 pCi/g dry wt)			
				Total beta	NA	Average = 44.7 pCi/g dry wt n = 39 (Range 30.8 - 56 4 pCi/g dry wt)			
			1982	Total alpha	NA	Average = 15.61 pCi/g dry wt n = 50 (Range 4.79 - 20.17 pCi/g dry wt)			
				Total beta	NA	Average = 42.1 pCi/g dry wt n = 50 (Range 25.3 - 46.8 pCi/g dry wt)			
The Metropolitan Sanitary District of Greater Chicago, 1984	Chicago - LASMA and HASMA	Lagoon sludge - "dried (non-volatile) material"	1982	Total alpha	NA	Average = 16.79 pCi/g dry wt n = 11 (Range 14.09 - 20.17 pCi/g dry wt)	Canberra alpha/ beta proportional counter	None specified	Fluctuating radioactivity concentrations
				Total beta	NA	Average - 41.5 pCi/g dry wt n = 11 (Range 31.8 - 45.7 pCi/g dry wt)			Note 1982 data reported in both 1982 and 1984 Annual Reports.

TABLE 1. SUMMARY OF RADIOACTIVITY LEVELS IN MUNICIPAL WASTEWATER TREATMENT SLUDGES (Continued)

Study	Location	Type of Sludge	Sample Period	Radio-nuclide	$T_{1/2}$	Radioactivity	Analytical Technique	Sources of Contamination	Comments/ Conclusions
The Metropolitan Sanitary District of Greater Chicago, 1984 (cont'd)			1983	Total alpha	NA	Average = 7.75 pCi/g dry wt n = 72 (Range 3.46 - 13.83 pCi/g dry wt)			
				Total beta	NA	Average = 45.5 pCi/g dry wt n = 74 (Range 37.5 - 63.3 pCi/g dry wt)			
			1984	Total alpha	NA	Average - 20.40 pCi/g dry wt n = 45 (Range 9.37 - 31.93 pCi/g dry wt)			
				Total beta	NA	Average = 47.5 pCi/g dry wt n = 46 (Range 27.9 - 69.7 pCi/g dry wt)			Overall means for 1978 to 1984 are. Total alpha = 14.22, n-288 Total beta = 49.4, n=305
Durham and Joshi, 1979	Hamilton Sewage Treatment Plant on Lake Ontario in Canada	Digester sludge, freeze dried for analysis	NR	^{144}Ce	284d	1248 ± 52 pCi/kg dry wt (ND in effluent)	Gamma-ray spectroscopy	Most radionuclides of natural origin or fallout from nuclear weapons testing except ^{51}Cr, ^{75}Se and ^{131}I which are used in nuclear medicine	^{51}Cr only radionuclide reaching Lake Ontario (from Hamilton's effluent). Most radioactivity removed with sludge during sewage treatment Concluded conc. of radionuclides in sludge are sufficiently low that agricultural use or landfilling do not represent radiation hazard.
				^{141}Ce	32.5d	145 ± 10 pCi/kg dry wt (ND in effluent)			
				^{226}Ra	1622y	1024 ± 106 pCi/kg dry wt (ND in effluent)			
				^{228}Th	1.9y	236 ± 14 pCi/kg dry wt (ND in effluent)			
				^{75}Se	120d	ND (ND in effluent)			
				^{51}Cr	27.8d	2705 ± 132 pCi/kg dry wt (1.24 ± 0.06 pCi/l in effluent)			
				^{131}I	8.06d	45 ± 10 pCi/kg dry wt (ND in effluent)			
				^{125}Sb	2.7y	143 ± 29 pCi/kg dry wt (ND in effluent)			
				^{7}Be	53.6d	2227 ± 139 pCi/kg dry wt (0.23 ± 0.05 pCi/l in effluent)			
				^{103}Ru	40d	229 ± 15 pCi/kg dry wt (ND in effluent)			

TABLE 1. SUMMARY OF RADIOACTIVITY LEVELS IN MUNICIPAL WASTEWATER TREATMENT SLUDGES (Continued)

Study	Location	Type of Sludge	Sample Period	Radio-nuclide	$T_{1/2}$	Radioactivity	Analytical Technique	Sources of Contamination	Comments/ Conclusions
Durham and Joshi, 1979 (cont'd)	Hamilton Sewage Treatment Plant (cont'd)			^{106}Ru	1y	951 ± 113 pCi/kg dry wt (ND in effluent)			
				^{137}Cs	30y	210 ± 17 pCi/kg dry wt (0.025 ± 0.005 pCi/l in effluent)			
				^{95}Zr	64d	254 ± 27 pCi/kg dry wt (ND in effluent)			
				^{95}Nb	35.1d	364 ± 22 pCi/kg dry wt (ND in effluent)			
	Dundas Sewage Treatment Plant on Lake Ontario in Canada	Digester sludge, freeze-dried for analysis	NR	^{144}Ce	284d	676 ± 53 pCi/kg dry wt (ND in effluent)	Gamma-ray spectroscopy	Same as for Hamilton STP	
				^{141}Ce	32.5d	ND (ND in effluent)			
				^{226}Ra	1622y	634 ± 97 pCi/kg dry wt (ND in effluent)			
				^{228}Th	1.9y	284 ± 20 pCi/kg dry wt (0.02 ± 0.01 pCi/l in effluent)			
				^{75}Se	120d	113 ± 12 pCi/kg dry wt (ND in effluent)			
				^{51}Cr	27.8d	ND (ND in effluent)			
				^{131}I	8.60d	185 ± 19 pCi/kg dry wt (ND in effluent)			
				^{125}Sb	2.7y	ND (ND in effluent)			
				^{7}Be	53.6d	827 ± 140 pCi/kg dry wt (ND in effluent)			
				^{103}Ru	40d	ND (ND in effluent)			
				^{106}Ru	1y	ND (ND in effluent)			

TABLE 1. SUMMARY OF RADIOACTIVITY LEVELS IN MUNICIPAL WASTEWATER TREATMENT SLUDGES (Continued)

Study	Location	Type of Sludge	Sample Period	Radionuclide	$T_{1/2}$	Radioactivity	Analytical Technique	Sources of Contamination	Comments/ Conclusions
Durham and Joshi, 1979 (cont'd)	Dundas Sewage Treatment Plant (cont'd)		NR	^{137}Cs	30y	100 + 20 pCi/kg (ND in effluent)			
				^{95}Zr	64d	ND (ND in effluent)			
				^{95}Nb	35.1d	ND (ND in effluent)			
Strong, 1973 as cited in Prichard et al., 1981	Denver	NR	1973	^{131}I	8d	57 pCi/kg wet wt	NR	NR	Conc. for each city based on a single sample. Other nuclides identified in the nine sludges sampled included ^{203}Hg, ^{232}Th, ^{137}Cs, ^{214}Bi, ^{106}Ru, and ^{40}K (conc. not given).
	Baltimore	NR	1973	^{131}I		54 pCi/kg wet wt	NR	NR	
	Chicago	NR	1973	^{131}I		46 pCi/kg wet wt	NR	NR	
	Houston	NR	1973	^{131}I		467 pCi/kg wet wt	NR	NR	
	Boston	NR	1973	^{131}I		ND	NR	NR	
	Philadelphia	NR	1973	^{131}I		ND	NR	NR	
	New York	NR	1973	^{131}I		142 pCi/kg wet wt	NR	NR	
	Los Angeles	NR	1973	^{131}I		674 pCi/kg wet wt	NR	NR	
	San Francisco	NR	1973	^{131}I		328 pCi/kg wet wt	NR	NR	
Williams, 1985	Colby, Wisconsin	Digested sludge (Contact stabilization activated sludge-aerobic digestion)	March, 1985	^{226}Ra	1622y	36.1 pCi/g dry wt (1085 pCi/l assuming 3% sludge solids)	Gamma-ray spectroscopy	Drinking water treatment-regeneration & backwashing ion exchange media	1 grab sample of sludge at each city (study gives conc. in ion exchange backwash, iron filter backwash, influent to sewage plants, effluent, & raw sludge
				Combined ^{226}Ra & ^{228}Ra	**	38.5 pCi/g dry wt (1157 pCi/l assuming 3% sludge solids)			Conclusions 1. Radium is concentrated in sludges. Occurs under aerobic and anaerobic conditions 2. Accumulation of Ra occurs in both fixed growth systems & suspended growth (activated sludge) systems. Additional conc. of Ra in sludge through digestion or heat treatment process. 3. Biological sludge will remove dissolved Ra from waste stream (e.g., St. Agnes, Fond du Lac & Lake Mills) as well as insoluble Ra very effectively. 4. Accumulation of Ra in sludge occurs at both low & high conc. in influent. 5. Radium in sludge appears to be in insoluble form in biological solids.
				Gross alpha Gross beta	NA NA	184.0 pCi/g 188.5 pCi/g	Alpha count Beta count		
	Congregation of St. Agnes, Wisconsin	Digested sludge (Conventional activated sludge-aerobic digestion)	March, 1985	^{226}Ra	1622y	14.6 pCi/g dry wt (440 pCi/l assuming 3% sludge solids)	Gamma-ray spectroscopy	Drinking water treatment-regeneration & backwashing ion exchange media	
				Combined ^{226}Ra & ^{228}Ra	**	24.9 pCi/g dry wt (750 pCi/l assuming 3% sludge solids)			
				Gross alpha Gross beta	NA NA	382.4 pCi/g 389.2 pCi/g	Alpha count Beta count		

TABLE 1. SUMMARY OF RADIOACTIVITY LEVELS IN MUNICIPAL WASTEWATER TREATMENT SLUDGES (Continued)

Study	Location	Type of Study	Sample Period	Radio Nuclide	$T_{1/2}$	Radioactivity	Analytical Technique	Sources of Contamination	Comments/ Conclusions
Williams, 1985, (cont'd)	Fond du Lac, Wisconsin	Heat treated sludge (Oxygen activated sludge - Zimpro Wet Air digestion)	March, 1985	^{226}Ra	1622y	22.4 pCi/g dry wt (673 pCi/l assuming 3% sludge solids)	Gamma-ray spectroscopy	Community water supply	
				Combined ^{226}Ra & ^{228}Ra	**	31.9 pCi/g dry wt (958 pCi/l assuming 3% sludge solids)			
				Gross alpha	NA	76.9 pCi/g	Alpha count		
				Gross beta	NA	83.9 pCi/g	Beta count		
	Juneau, Wisconsin	Digested sludge (Extended aeration activated sludge - aerobic digestion)	June, 1985	^{226}Ra	1622y	17.3 pCi/g dry wt (571 pCi/l with 3.3% solids, actual)	Gamma-ray spectroscopy	Drinking water treatment-backwashing of iron filter (note--backwash normally not discharged to sanitary sewage lines-only done for this project)	
				Combined ^{226}Ra & ^{228}Ra	**	20.9 pCi/g dry wt (692 pCi/l with 3.3% solids, actual)			
				Gross alpha	NA	119.6 pCi/g	Alpha count		
				Gross beta	NA	96.8 pCi/g	Beta count		
	Lake Mills, Wisconsin	Digested sludge (Rotating biological contractors - anaerobic digestion)	February, 1985	^{226}Ra	1622y	26.7 pCi/g dry wt (1120 pCi/l with 4.2% solids, actual)	Gamma-ray spectroscopy	Community water supply	
				Combined ^{226}Ra & ^{228}Ra	**	31.9 pCi/g dry wt (1565 pCi/l with 4.2% solids, actual)			
				Gross alpha	NA	109.8 pCi/g	Alpha count		
				Gross beta	NA	79.8 pCi/g	Beta count		

TABLE 1. SUMMARY OF RADIOACTIVITY LEVELS IN MUNICIPAL WASTEWATER TREATMENT SLUDGES (Continued)

Study	Location	Type of Sludge	Sample Period	Radio-nuclide	$T_{1/2}$	Radioactivity	Analytical Technique	Sources of Contamination	Comments/ Conclusions
Texas Department of Health, 1986	Webster Wastewater Treatment Plant in Webster, Texas	Dried processed sludge	3-22-83	^{241}Am	432y	$6.61 \times 10^{-6} \pm 2.40 \times 10^{-6}$ µCi/g	Gamma scan	Gulf Nuclear, Inc.	Raw data from lab sheets. Webster WWTP is a contact stabilization process (modification of activated sludge).
				^{137}Cs	30y	$2.79 \times 10^{-5} \pm 1.33 \times 10^{-6}$ µCi/g			
				^{131}I	8d	$1.63 \times 10^{-6} \pm 3.74 \times 10^{-7}$ µCi/g			
				^{192}Ir	74d	$9.73 \times 10^{-6} \pm 1.20 \times 10^{-6}$ µCi/g			
				^{153}Gd	242d	$<1.07 \times 10^{-6}$ µCi/g			
				Gross alpha	NA	$1.0 \times 10^{-5} \pm 7.3 \times 10^{-6}$ µCi/g	Alpha count		
				Gross beta	NA	$5.8 \times 10^{-5} \pm 1.5 \times 10^{-5}$ µCi/g	Beta count		
		Surface sludge	3-22-83	^{241}Am	432y	$1.90 \times 10^{-6} \pm 8.25 \times 10^{-7}$ µCi/g	Gamma scan		
				^{137}Cs	30y	$1.89 \times 10^{-5} \pm 1.04 \times 10^{-6}$ µCi/g			
				^{192}Ir	74d	$1.58 \times 10^{-5} \pm 1.98 \times 10^{-6}$ µCi/g			
				^{131}I	8d	$<6.08 \times 10^{-6}$ µCi/g			
		Surface sludge	3-22-83	^{241}Am	432y	$2.60 \times 10^{-5} \pm 8.49 \times 10^{-6}$ µCi/g	Gamma scan		
				^{137}Cs	30y	$6.18 \times 10^{-5} \pm 2.70 \times 10^{-6}$ µCi/g			
				^{192}Ir	74d	$2.57 \times 10^{-5} \pm 2.98 \times 10^{-6}$ µCi/g			
				^{131}I	8d	$<1.16 \times 10^{-6}$ µCi/g			
		Process sludge	3-22-83	^{241}Am	432y	$1.95 \times 10^{-5} \pm 6.66 \times 10^{-6}$ µCi/g	Gamma scan		
				^{137}Cs	30y	$5.25 \times 10^{-5} \pm 2.82 \times 10^{-6}$ µCi/g			
				^{192}Ir	74d	$3.23 \times 10^{-5} \pm 4.11 \times 10^{-6}$ µCi/g			
				^{131}I	8d	$<1.77 \times 10^{-6}$ µCi/g			

TABLE 1. SUMMARY OF RADIOACTIVITY LEVELS IN MUNICIPAL WASTEWATER TREATMENT SLUDGES (Continued)

Study	Location	Type of Sludge	Sample Period	Radio-nuclide	$T_{1/2}$	Radioactivity	Analytical Technique	Sources of Contamination	Comments/ Conclusions
Texas Department of Health, 1986 (cont'd)	Webster Wastewater Treatment Plant in Webster, Texas (cont'd)	Process sludge	3-28-83	^{241}Am	432y	$1.38 \times 10^{-5} \pm 5.0 \times 10^{-6}$ μCi/g	Gamma scan		
				^{137}Cs	30y	$5.36 \times 10^{-5} \pm 3.2 \times 10^{-6}$ μCi/g			
		Sludge	3-31-83	^{241}Am	432y	$9.8 \times 10^{-6} \pm 3.4 \times 10^{-6}$ μCi/g	Gamma scan		
				^{137}Cs	30y	$3.1 \times 10^{-5} \pm 2.0 \times 10^{-6}$ μCi/g			
				^{192}Ir	74d	$1.1 \times 10^{-5} \pm 1.6 \times 10^{-6}$ μCi/g			
				^{153}Gd	242d	1.5×10^{-6} μCi/g			
		Sludge bin (soil) (2 samples)	8-17-83	^{137}Cs	30y	$8.0 \times 10^{-5} \pm 5.0 \times 10^{-6}$ μCi/g	Gamma scan		
				^{192}Ir	74d	$1.40 \times 10^{-4} \pm 9.7 \times 10^{-6}$ μCi/g			
				^{241}Am	432y	8.9×10^{-6} μCi/g			
				^{131}I	8d	$6.2 \times 10^{-6} \pm 1.1 \times 10^{-6}$ μCi/g			
				^{153}Gd	242d	·4.4×10^{-6} μCi/g			
				Gross alpha	NA	$2.7 \times 10^{-5} \pm 7.3 \times 10^{-6}$ μCi/g	Alpha count		
				Gross beta	NA	$3.65 \times 10^{-4} \pm 1.9 \times 10^{-5}$ μCi/g	Beta count		
			8-17-83	^{137}Cs	30y	$7.5 \times 10^{-5} \pm 4.4 \times 10^{-6}$ μCi/g	Gamma scan		
				^{192}Ir	74d	$3.7 \times 10^{-5} \pm 2.7 \times 10^{-6}$ μCi/g			
				^{241}Am	432y	6.5×10^{-6} μCi/g			
				^{131}I	8d	$6.4 \times 10^{-6} \pm 8.3 \times 10^{-7}$ μCi/g			
				^{153}Gd	242d	·3.0×10^{-6} μCi/g			
				^{40}K	1.28×10^{9}y	$7.9 \times 10^{-6} \pm 3.5 \times 10^{-6}$ μCi/g			
				Gross alpha	NA	$2.4 \times 10^{-5} \pm 6.8 \times 10^{-6}$ μCi/g	Alpha count		
				Gross beta	NA	$9.5 \times 10^{-5} \pm 1.2 \times 10^{-5}$ μCi/g	Beta count		

TABLE 1. SUMMARY OF RADIOACTIVITY LEVELS IN MUNICIPAL WASTEWATER TREATMENT SLUDGES (Continued)

Study	Location	Type of Sludge	Sample Period	Radionuclide	$T_{1/2}$	Radioactivity	Analytical Technique	Sources of Contamination	Comments/ Conclusions
Texas Department of Health, 1986 (cont'd)	Webster Wastewater Treatment Plant in Webster, Texas (cont'd)	Sludge on floor of drier building	9-29-83	^{137}Cs	30y	$4.87 \times 10^{-5} \pm 1.7 \times 10^{-6}$ µCi/g	Gamma scan		
				^{192}Ir	74d	$4.1 \times 10^{-6} \pm 4 \times 10^{-7}$ µCi/g			
				^{241}Am	432y	LLDL 7.4×10^{-6} µCi/g			
				^{7}Be	53d	LLDL 1.12×10^{-5} µCi/g			
				^{60}Co	5.3y	LLDL 8×10^{-7} µCi/g			
				^{129}I	1.59×10^{7}y	LLDL 8.87×10^{-5} µCi/g			
				^{131}I	8d	LLDL 2.2×10^{-6} µCi/g			
				^{3}H	12.3y	$<5.0 \times 10^{-7}$ µCi/ml	Beta count		
				^{14}C	5730y	$<2.0 \times 10^{-6}$ µCi/g			
		Sludge on drier belt	9-29-83	^{226}Ra	1622y	$7.23 \times 10^{-5} \pm 7.8 \times 10^{-6}$ µCi/g	Gamma scan		
				^{241}Am	432y	LLDL 7.3×10^{-6} µCi/g			
				^{7}Be	53d	LLDL 1.39×10^{-5} µCi/g			
				^{137}Cs	30y	LLDL 1.0×10^{-6} µCi/g			
				^{60}Co	5.3y	LLDL 1.0×10^{-6} µCi/g			
				^{129}I	1.59×10^{7}y	LLDL 1.19×10^{-4} µCi/g			
				^{131}I	8d	LLDL 2.7×10^{-6} µCi/g			
				^{192}Ir	74d	LLDL 1.4×10^{-6} µCi/g			
				^{3}H	12.3y	$<5.0 \times 10^{-7}$ µCi/ml	Beta count		
				^{14}C	5730y	$<2.0 \times 10^{-5}$ µCi/g			

TABLE 1. SUMMARY OF RADIOACTIVITY LEVELS IN MUNICIPAL WASTEWATER TREATMENT SLUDGES (Continued)

Study	Location	Type of Sludge	Sample Period	Radio-nuclide	$T_{1/2}$	Radioactivity	Analytical Technique	Sources of Contamination	Comments/ Conclusions
Texas Department of Health, 1986 (cont'd)	Webster Wastewater Treatment Plant in Webster, Texas (cont'd)	Dried sludge	1-14-86	^{228}Ac	6h	$1.0 \times 10^{-6} \pm 2 \times 10^{-7}$ μCi/g	Gamma scan		
				^{241}Am	432y	$6 \times 10^{-7} \pm 5 \times 10^{-7}$ μCi/g			
				^{7}Be	53d	$2.8 \times 10^{-6} \pm 4 \times 10^{-7}$ μCi/g			
				^{137}Cs	30y	$1.9 \times 10^{-6} \pm 2 \times 10^{-7}$ μCi/g			
				^{131}I	8d	$2.9 \times 10^{-6} \pm 2 \times 10^{-7}$ μCi/g			
				^{192}Ir	74d	$2.2 \times 10^{-6} \pm 2 \times 10^{-7}$ μCi/g			
				^{40}K	1.28×10^{7}y	$4.1 \times 10^{-6} \pm 8 \times 10^{-7}$ μCi/g			
				^{212}Pb	10.6h	$4 \times 10^{-7} \pm 2 \times 10^{-7}$ μCi/g			
				^{214}Pb	26.8m	$2 \times 10^{-7} \pm 1 \times 10^{-7}$ μCi/g			
				Gross beta	NA	$1.3 \times 10^{-5} \pm 5 \times 10^{-6}$ μCi/g	Beta count		
	Sims Bayou WWTP	Heat treated sludge	9-29-83	^{228}Ac	6h	$1.8 \times 10^{-6} \pm 5 \times 10^{-7}$ μCi/g	Gamma scan		
				^{131}I	8d	$8.0 \times 10^{-6} \pm 9 \times 10^{-7}$ μCi/g			Same facility investigated by Prichard et al., 1981.
				^{14}C	5730y	5.0×10^{-6} μCi/g	Beta count		
		Dried sludge	9-29-83	^{131}I	8d	$4.03 \times 10^{-5} \pm 4.0 \times 10^{-6}$ μCi/g	Gamma scan		
				^{3}H	12.3y	$7.9 \times 10^{-7} \pm 8 \times 10^{-8}$ μCi/ml	Beta count		
				^{14}C	5730y	$4.7 \times 10^{-6} \pm 4 \times 10^{-7}$ μCi/g			
New York State Department of Health, 1986	Tonawanda, NY	Sludge, Tonawanda Sewage Treatment Plant (TSTP)	5-11-84	^{241}Am	432y	5.2 ± 1.2 pCi/g	FIDLER (Field Instrument for Detection of Low Energy Radiation) scintillation probe	EAD Metallurgical, Inc., producer of foil elements in smoke detectors	Radiological monitoring performed following inspection at EAD on 4-26-84 indicating contamination of sewer line. Data from computer printout and draft report by NY Dept. of Health.
		Activated Sludge, TSTP	5-17-84	^{241}Am		12 0 ± 3 0 pCi/g			
		Sludge, bottom of settling tank, TSTP	5-17-84	^{241}Am		5 8 ± 1.5 pCi/g			

TABLE 1. SUMMARY OF RADIOACTIVITY LEVELS IN MUNICIPAL WASTEWATER TREATMENT SLUDGES (Continued)

Study	Location	Type of Sludge	Sample Period	Radio-nuclide	$T_{1/2}$	Radioactivity	Analytical Technique	Sources of Contamination	Comments/ Conclusions
New York State Department of Health, 1986 (cont'd)	Tonawanda, NY (cont'd)	Sludge after Zimbro Process, TSTP	5-17-84	^{241}Am	432y	5.4 ± 1.4 pCi/g			
		Sludge before Zimbro Process, TSTP	6-19-84	^{241}Am		4.5 ± 1.6 pCi/g			
		Sludge, TSTP	6-84 to 12-84	^{241}Am		$\bar{x}$ = 3.5 pCi/g, n = 23 (Range 1.8 - 5.0 pCi/g)			$\bar{x}$ excludes one sample of 100 pCi/g taken 8-07-84.
		Sludge, TSTP	1-85 to 1-86	^{241}Am		$\bar{x}$ = 2.5 pCi/g, n = 26 (Range 0.9 - 12 pCi/g)			
		Ash, Incinerator #2, TSTP	4-26-84	^{241}Am		500 ± 150 pCi/g			Contaminated ash conc. of ^{241}Am ranged from 160 - 750 pCi/g, current ash contains low conc. of ^{241}Am
		Ash from landfill (2 years old)	4-26-84	^{241}Am		300 ± 100 pCi/g			
		Truck bed clinker (from incinerated ash)	5-07-84	^{241}Am		420 ± 100 pCi/g			
		Ash, Incinerator #2, TSTP (2 samples)	5-07-84	^{241}Am		180 ± 40 pCi/g			
			5-07-84	^{241}Am		750 ± 190 pCi/g			
		Clinker from Incinerator #2, TSTP	5-07-84	^{241}Am		620 ± 160 pCi/g			
		Ash from TSTP	5-84 to 12-84	^{241}Am		$\bar{x}$ = 11.7 pCi/g, n = 25 (Range 6.0 - 22 pCi/g)			
		Ash from TSTP	1-85 to 1-86	^{241}Am		$\bar{x}$ = 6.1 pCi/g, n = 24 (Range 2.2 - 23 pCi/g)			$\bar{x}$ excludes 2 samples of 63 pCi/g and 80 pCi/g taken 2-07-85 and 12-26-85, respectively.
		Ash from Niagara landfill (2 samples)	5-16-84	^{241}Am		10 ± 3 pCi/g			
	Grand Island, NY	Pressed sludge	8-84 to 12-84	^{241}Am		$\bar{x}$ = 83.7 pCi/g, n = 12 (Range 57 - 120 pCi/g)	Unspecified	NRD (manufacturer of Am-241 foils) in Grand Island, NY	

TABLE 1. SUMMARY OF RADIOACTIVITY LEVELS IN MUNICIPAL WASTEWATER TREATMENT SLUDGES (Continued)

Study	Location	Type of Sludge	Sample Period	Radio-nuclide	$T_{1/2}$	Radioactivity	Analytical Technique	Sources of Contamination	Comments/ Conclusions
New York State Department of Health, 1986 (cont'd)	Grand Island, NY (cont'd)	Pressed sludge (cont'd)	1-85 to 6-85	^{241}Am	432y	$\bar{x}$ = 39.7 pCi/g, n = 12 (Range 33 - 53 pCi/g)			
			7-85 to 11-85	^{241}Am		$\bar{x}$ = 24.8 pCi/g, n = 10 (Range 15 - 43 pCi/g)			
			1-15-86	^{241}Am		5.7 ± 1.5 pCi/g			
		Unpressed sludge	9-84 to 11-84	^{241}Am		$\bar{x}$ = 76.8 pCi/g, n = 5 (Range 56 - 100 pCi/g)			
		Digester sludge	10-10-84	^{241}Am		17 ± 6 pCi/g			

NA = Not Applicable
ND = Not Detected
NR = Not Reported

mCi = millicuries=1 x 10^{-3} curies
μCi = microcuries = 1 x 10^{-6} curies
pCi = picocuries - 1 x 10^{-12} curies
fCi = femtocuries = 1 x 10^{-15} curies

LLDL = Lower Limit Detectable

* $T_{1/2}$ for ^{239}Pu = 2.411 x 10^4 years, $T_{1/2}$ for ^{240}Pu = 6537 years

** $T_{1/2}$ for ^{228}Ra = 5.75 years limited to those which contained data on concentrations of radionuclides in sludges from the United States and Canada. Related studies of sludge from foreign countries and studies which contained information on concentrations of radionuclides in wastewater treatment effluents or in soils treated with sludges were not included in Table 1. However, these studies are discussed in relation to the results of sludge studies when appropriate.

One source of radioactivity in municipal wastewater sludge is medical treatment and research. Radionuclides are released to the sewage system through isotope releases in handling and through excretion by patients. In sludges from a Texas sewage treatment plant serving nine institutions with active nuclear medicine programs, Prichard et al. (1981) found that iodine-131 (^{131}I) dominated the gamma spectra. Concentrations of ^{131}I ranged from approximately 7 pCi/g to 180 pCi/g in flash dried sludge and approximately 1 pCi/g to 27 pCi/g in vacuum dried sludge (an earlier step in the sludge drying process). The authors noted that none of the activity was lost in the flash drying process. The ratio of ^{131}I in sludge to ^{131}I in liquid effluent from the plant was 1:3.3 indicating that some, though not all, of the ^{131}I was retained in the sludge. The authors also noted that the sludge would not be expected to produce a significant population dose when used as a soil conditioner because the time required to process the sludge and to plant and harvest a crop in treated soil greatly exceeded the 8-day half-life of ^{131}I.

Strong (1973, as cited in Prichard et al., 1981) reported ^{131}I concentrations for ten major U.S. cities (Table 1). These concentrations ranged from levels below detection to 0.674 pCi/g of wet sludge. The amount of moisture in these sludges was not reported, making comparison to Prichard's results difficult. A Swedish study by Erlandsson and Mattsson (1978) detected ^{131}I in all samples from a sewage treatment plant serving an area containing one hospital and where most patients treated at that hospital lived in the district. Concentrations ranged from 0.03 pCi/g to 0.12 pCi/g of ^{131}I in digested sludge (23 percent dry substance). Moss (1973) found that ^{131}I and technicium-99m (^{99m}Tc) constituted 95 percent of the total activity (13-15 pCi/l) in effluent from a sewage plant in Pennsylvania. Sodd et al. (1975) sampled effluents from a sewage treatment plant near

Cincinnati, Ohio, and found measurable concentrations of ^{99m}Tc and ^{131}I. Concentrations of ^{99m}Tc ranged from 4 pCi/l to 217 pCi/l; concentrations of ^{131}I ranged from 18 pCi/l to 134 pCi/l. The study by Erlandsson and Mattsson presented data from outside the United States and Canada, and the studies of Moss (1973) and Sodd et al. (1975) did not present data for sludges. Therefore, these studies were not included in Table 1.

Durham and Joshi (1979) sampled digested sludge from the sewage treatment plants in Hamilton and Dundas, Ontario, on the western shore of Lake Ontario. This study was performed in response to concern raised by the Great Lakes International Joint Commission (GLIJC) that the discharge of radionuclides might affect the water quality of the Great Lakes. Results of this study are also presented in the Great Lakes Water Quality Seventh Annual Report (GLIJC, 1979). Fourteen radionuclides were detected in sludges from the two treatment plants. Most radionuclides were present at very low concentrations. The authors noted that most were of natural origin or resulted from fallout of nuclear weapons testing. However, three of the radionuclides detected -- chromium-51 (^{51}Cr), selenium-75 (^{75}Se), and ^{131}I -- were used locally in nuclear medicine. Thirteen radionuclides were detected in sludge from the Hamilton plant. The radionuclides with the highest concentrations were ^{51}Cr, 2.705 $\pm$ 0.132 pCi/g; beryllium-7 (^{7}Be), 2.227 $\pm$ 0.139 pCi/g; cerium-144 (^{144}Ce), 1.248 $\pm$ 0.052 pCi/g; radium-226 (^{226}Ra), 1.024 $\pm$ 0.106 pCi/g; and ruthenium-106 (^{106}Ru), 0.951 $\pm$ 0.113 pCi/g. All other radionuclides were below 0.5 pCi/g. Seven radionuclides were detected in sludge from the Dundas plant. The highest concentrations were ^{7}Be, 0.827 $\pm$ 0.140 pCi/g; ^{144}Ce, 0.676 $\pm$ 0.053 pCi/g; and ^{226}Ra, 0.634 $\pm$ 0.097 pCi/g. ^{51}Cr, ^{7}Be, and cesium-137 (^{137}Cs) were detected in effluent from the Hamilton plant and thorium-228 (^{228}Th) was detected in effluent from the Dundas plant. ^{51}Cr was the only radionuclide found to reach Lake Ontario.

The Metropolitan Sanitary District of Greater Chicago has conducted a program of radiological monitoring of sludges from its sewage lagoons since 1978. Data were obtained for the years of 1978 through 1984 (Metropolitan Sanitary District of Greater Chicago, 1979, 1982, and 1984). Radioactivity was analyzed only as "total alpha" and "total beta" concentrations. Annual means were computed from data presented in annual reports of the Metropolitan

Sanitary District and are presented in Table 1. Concentrations were shown to fluctuate within a particular year and between years, but no exceptional surges in radioactivity levels were recorded. For example, between 1978 and 1984, the total alpha concentrations ranged from 3.89 to 39.12 pCi/g dry wt. and total beta concentrations ranged from 25.3 to 126 pCi/g dry wt.

The organic fertilizer product "Nu Earth", distributed free to the public by the Metropolitan Sanitary District in Stickney, Illinois was analyzed for radioactivity in 1974 and 1975 (Alberts et al., 1974; Alberts and Wahlgren, 1977). The organic fertilizer consisted of air dried sediment from an Imhoff Process. The radionuclides analyzed were plutonium-239,240 ($^{239,240}Pu$), americium-241 (^{241}Am), and ^{137}Cs. Sludge samples collected in Summer, 1974 contained 21.4 fCi/g dry wt. of $^{239,240}Pu$ and 3.3 fCi/g dry wt. of ^{241}Am. Samples collected the following year in Spring, 1975 contained 12.8 fCi/g dry wt. of $^{239,240}Pu$ and 1.5 fCi/g dry wt. of ^{241}Am. The ratios of $^{239,240}Pu/^{241}Am$ for the 1974 and 1975 samples were 6.5 and 8.5, respectively. These ratios were considered to be more representative of the soil environment than of lake sediment. Therefore, the source of $^{239,240}Pu$ and ^{241}Am was thought to be runoff from the metropolitan area rather than transport from sediments of Lake Michigan introduced through drinking water (Alberts and Wahlgren, 1977). ^{137}Cs and $^{239,240}Pu$ were detected in a November, 1974 sample at concentrations of 1.08 pCi/g dry wt. and 21.5 fCi/g dry wt., respectively (Alberts et al., 1974).

Another source of radionuclides in sludge is drinking water containing elevated concentrations of radium. These elevated concentrations of radium are found in areas of the county where the water is taken from wells in high radium formations such as granite. Many of the cities in these areas are required under the Safe Drinking Water Act to treat their drinking water to reduce radium concentrations to acceptable levels (5 pCi/l) (40 CFR 141). In the water treatment process, some systems such as ion exchange columns require backflushing to clean the radium removal columns, and water from backflushing is often discharged to the sewers. Other water treatment systems produce sludge which may be discharged to the sewers. Thus, radium can enter the sanitary sewers through untreated drinking water or through discharge of backflushed water or sludges from water treatment systems.

Williams (1985) recently completed a study which analyzed sludge from municipal sewage treatment plants in several Wisconsin cities where drinking water was known to contain elevated concentrations of ^{226}Ra and ^{228}Ra. Concentrations of ^{226}Ra in sludges from the five communities sampled ranged from 14.6 pCi/g dry wt. to 36.1 pCi/g dry wt., and combined concentrations of ^{226}Ra and ^{228}Ra ranged from 20.9 pCi/g dry wt. to 38.5 pCi/g dry wt. Although the study is based on limited sampling (one sample per city), the author made several interesting observations. The results indicated that activated sludge effectively adsorbed ^{226}Ra and ^{228}Ra. Removal of radium occurred at both high and low influent concentrations and was effective for both soluble and insoluble forms of radium. Accumulation of radium in sludge occurred under aerobic and anaerobic conditions and in fixed growth and suspended growth systems. Also, accumulation of radium in sludge was observed both in cities in Wisconsin that discharge backflush waters from water treatment and in cities where the radium was from the untreated community water supply itself.

In 1984, the U.S. Nuclear Regulatory Commission (NRC) issued notices to state radiation control agencies and to NRC materials licensees raising concern for the potentially significant problem of discharging radionuclides into sanitary sewage systems (Nussbaumer, 1984; Grace, 1984). In these notices, NRC cited three incidents of contamination of sewage sludge attributable to releases from licensed industrial users of radionuclides. Two incidents of contamination of sludge with ^{241}Am from companies producing foils for smoke alarms occurred in New York State. Sludge data for these incidents were obtained from the New York State Department of Health (1986). The sewage treatment facilities involved were the Tonawanda Sewage Treatment Plant which incinerates its sludge to reduce the volume for landfilling and the Grand Island Sewage Treatment Plant which landfills its sludge.

The concentrations reported in contaminated ash from Tonawanda ranged from 180 $\pm$ 40 pCi/g ash to 750 $\pm$ 190 pCi/g ash between April 17 and May 7, 1984. Concentrations measured recently have dropped to approximately 10 $\pm$ 2 pCi/g ash. Sampling of sludges did not begin until May 11, 1984, approximately two weeks after the initiation of ash sampling. At this time, concentrations of ^{241}Am in ash had dropped from the aforementioned elevated concentrations to 13 pCi/g on May 11, 1984. The corresponding sludge concentration on this date was 5.2 pCi/g. Presumably the lower concentrations indicate that the release of ^{241}Am to the sewage system had ceased. The mean concentrations in sludge were 3.5 pCi/g for the last half of 1984 and 2.5 pCi/g for 1985. Concentrations of ^{241}Am in sludge at Grand Island were as high as 120 $\pm$ 30 pCi/g in 1984 and have dropped progressively since that time. Sludge samples taken at Grand Island in the last half of 1985 averaged 24.8 pCi/g and a sample taken in January of 1986 contained 5.7 $\pm$ 1.5 pCi/g.

A third incident of sludge contamination reported by NRC (Grace, 1984; Nussbaumer, 1984) occurred in Oak Ridge, Tennessee, where the sludge was being landspread as fertilizer at a U.S. Department of Energy reforestation project. The apparent source of contamination was a state licensee who occasionally discharged a few thousand gallons of liquid per day into the sanitary system containing cobalt-60 (^{60}Co) at 66-110 disintigrations per minute (dpm) per milliliter. The principal sludge contaminant was ^{60}Co, although ^{137}Cs and ^{134}Cs were also identified. Sludge samples contained 10 to 100 pCi/g gross activity (20,000 - 200,000 dpm/kg) (Grace, 1984; Nussbaumer, 1984) No further information was available on sludge concentrations and, therefore, this study was not included in Table 1. However, Oakes et al. (1984a and 1984b) performed a study examining the concentrations of radionuclides in the soils at the 65-acre landspreading site and analyzing potential pathways of exposure. Concentrations in the top three inches of soil were considered to be most representative of the sludge applied. Concentrations of ^{60}Co ranged from 0.08 to 44.8 pCi/g dry wt. with a mean of 2.96 pCi/g. Concentrations of ^{137}Cs ranged from 0.09 to 15.4 pCi/g with a mean of 1.72 pCi/g. Mean concentrations for other radionuclides in the soil were as follows: strontium-90 (^{90}Sr), 0.6 pCi/g; uranium-234 (^{234}U), 1.12 pCi/g; ^{235}U, 0.09 pCi/g; ^{238}U, 0.47 pCi/g; ^{238}Pu, 8 x 10^{-3} pCi/g; and ^{239}Pu, 0.039 pCi/g. The authors estimated that a total of 170 mCi of activity was present in the top three-inch layer of soil at the 65-acre site, 69 percent of which was contributed by ^{60}Co and ^{137}Cs, 23 percent by ^{234}U and ^{90}Sr, and 8 percent by other radionuclides including ^{238}U, ^{235}U, ^{239}Pu, and ^{238}Pu.

The Texas Department of Health (1986) provided raw data on radionuclide concentrations in sludge samples taken during 1983 and 1986 at the Webster and Sims Bayou Wastewater Treatment Plants. The Webster Treatment Plant receives sanitary waste from the Gulf Nuclear, Inc. facility, and an environmental survey was performed in 1983 to monitor the release of radionuclides from this facility. The laboratory analysis sheets noted suspected contaminants as being iridium-192 (^{192}Ir), gadolinium-153 (^{153}Gd), ^{137}Cs, ^{241}Am, and ^{131}I. Data were also available for the Sims Bayou Treatment Plant. Concentrations at the two facilities were reported for a variety of radionuclides and are presented in Table 1.

Mumma and coworkers sampled sludges from cities in central New York State (Mumma et al., 1983) and from cities across the United States (Mumma et al., 1984). Radioactivity was measured as gamma emission, and data were reported as counts per minute (cpm) above background. However, no data were presented on specific radionuclides or energies. Both studies concluded that gamma emission from sludge did not present a problem. Since the data were not in a form which could easily be compared to the estimated limits or standards, results of these studies are not presented in Table 1. The results are, however, presented for reference in Tables C-1 and C-2 of Appendix C.

SUMMARY

Results of the literature search and telephone survey identified nine sources containing data on radioactivity concentrations in municipal wastewater treatment sludges (Table 1). The obtained data varied widely with respect to purpose of collection, types of sludges sampled, number of samples, and radionuclides analyzed. In many cases, these factors made direct comparison of the data difficult or impossible. Review of the data indicated that some radionuclides were reported more often than others. Generally, the radionuclides most frequently reported were those associated with suspected or known contamination problems.

The four radionuclides reported most often in the available data were ^{131}I, ^{226}Ra, ^{241}Am, and ^{137}Cs. Of these four, three originated from human use of radionuclides (^{131}I, ^{241}Am, and ^{137}Cs) and one (^{226}Ra) originated from natural sources. ^{131}I, which is widely used in nuclear medicine, was reported in sludge from 11 sewage treatment plants (approximately 20 samples). Reported concentrations of ^{131}I ranged from 0.045 to 180 pCi/g. ^{226}Ra was reported in sludge from eight sewage treatment plants (approximately eight samples). For four of the plants, sludge was analyzed specifically for radium due to its natural presence at high concentrations in drinking water. Concentrations of ^{226}Ra ranged from 0.634 to 72.3 pCi/g. ^{241}Am was reported at four sewage treatment plants (approximately 164 samples of sludge and ash from sludge incineration). At two locations where approximately 150 of the samples were taken, ^{241}Am had been released to the sewage systems by manufacturers of foils used in smoke detectors. Concentrations in sludge ranged from 0.002 to 100 pCi/g while concentrations in contaminated ash were as high as 750 pCi/g. Concentrations of ^{137}Cs were reported in sludges from four sewage treatment plants (approximately 14 samples). Concentrations ranged from 0.10 to 80 pCi/g. The apparent sources of ^{137}Cs were the Gulf Nuclear facility and fallout.

REFERENCES

Alberts, J. J., and M. A. Wahlgren. 1977. Concentrations of Pu-239,240 and Am-241 in Drinking Water and Organic Fertilizer. Health Physics 32:295-297.

Alberts, J. J., P. J. Jehn, D. M. Nelson, J. S. Marshall, and M. A. Wahlgren. 1974. Effect of Municipal Treatment Processes on Pu-239,240 and Cs-137. In: Radiological and Environmental Research Division Annual Report, Ecology, January through December 1974. ANL-75-3, Part III. Argonne National Laboratory, Argonne, IL.

Durham, R. W., and S. R. Joshi. 1979. Radionuclide Concentrations in Two Sewage Treatment Plants on Western Lake Ontario, Canada. J. Radioanalytical Chem. 54 (1-2):367-370.

Erlandsson, B., and S. Mattsson. 1978. Medically Used Radionuclides in Sewage Sludge. Water, Air, Soil Pollution 9:199-206.

Grace, J. N. 1984. Information Notice No. 84-94 to All NRC Materials Licensees Other than Licensees that Use Sealed Sources Only. Regarding Reconcentration of Radionuclides Involving Discharges into Sanitary Sewage Systems Permitted under 10 CFR 20.303. Issued by J. N. Grace, Director, Div. of Quality Assurance, Safeguards and Inspection Programs, Office of Inspection and Enforcement, U.S. Nuclear Regulatory Commission, Washington, DC, December 21, 1984.

Great Lakes International Joint Commission. 1979. Great Lakes Water Quality, Seventh Annual Report, Appendix D, Annual Report of the Radioactivity Subcommittee. Great Lakes Regional Office International Joint Commission, Windsor, Ontario.

Metropolitan Sanitary District of Greater Chicago. 1979. Research and Development Department 1979 Annual Report. Chicago, IL.

Metropolitan Sanitary District of Greater Chicago. 1982. Research and Development Department 1982 Annual Report. Chicago, IL.

Metropolitan Sanitary District of Greater Chicago. 1984. Research and Development Department 1984 Annual Report. Chicago, IL.

Moss, C. E. 1973. Control of Radioisotope Releases to the Environment from Diagnostic Isotope Procedures. Health Physics 25:197-198.

Mumma, R. O., D. R. Raupach, J. P. Waldman, J. H. Hotchkiss, W. H. Gutenmann, C. A. Bache, and D. J. Lisk. 1983. Analytical Survey of Elements and Other Constituents in Central New York State Sewage Sludges. Arch. Environ. Contam. Toxicol. 12:581-587.

Mumma, R. O., D. R. Raupach, J. P. Waldman, S. S. C. Tong, M. L. Jacobs, J. G. Babish, J. H. Hotchkiss, P. C. Wszolek, W. H. Gutenmann, C. A. Bache, and D. J. Lisk. 1984. National Survey of Elements and Other Constituents in Municipal Sewage Sludges. Arch. Environ. Contam. Toxicol. 13:75-83.

New York State Department of Health. 1986. Data on Americium-241 Levels in Sludges from the Tonawanda and Grand Island Sewage Treatment Plants Following Contamination From Industries Using Americium-241 in Smoke Detector Foils. Obtained through personal communications with Dr. Rimawi, NY Department of Health, Albany, New York.

Nussbaumer, D. A., 1984*. Memorandum to All Agreement States regarding Sewer Sludge Contamination, from D. A. Nussbaumer, Assist. Dir. State Agreements Program, Office of State Programs, U.S. Nuclear Regulatory Commission.

Oakes, T. W., H. M. Braunstein, K. L. Daniels, W. F. Ohnesorge, J. T. Kitchings, and W. A. Alexander. 1984a. Report on the Oak Ridge Sewage Sludge Land-Farming Experience. Part I - Data Presentation. ORNL-6062/P1. Department of Environmental Management, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Oakes, T. W., W. F. Ohnesorge, K. L. Daniels, H. M. Braunstein, J. T. Kitchings, and W. A. Alexander. 1984b. Report on the Oak Ridge Sewage Sludge Land-Farming Experience. Part II - Pathways Analysis. ORNL-6062/P2. Department of Environmental Management, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

Prichard, H. M., T. F. Gessell, and E. Davis. 1981. Iodine-131 Levels in Sludge and Treated Municipal Wastewaters Near a Large Medical Complex. Am. J. Public Health 71(1):47-52.

Sodd, V. J., R. J. Velten, and E. L. Saenger. 1975. Concentrations of the Medically Useful Radionuclides, Technetium-99m and Iodine-131 at a Large Metropolitan Waste Water Treatment Plant. Health Physics 28:355-359.

Strong, A. B. 1973. Municipal Sludge Samples. Memorandum to Charles Weaver from Chief, Analytical Services Branch, Eastern Environmental Research Laboratory, U.S. Environmental Protection Agency, March 22, 1973. (As cited in Prichard et al., 1981).

Texas Department of Health. 1986. Data on Radioactivity in Municipal Wastewater Treatment Plants; Webster Plant, 1983 and 1986 and Sims Bayou Plant, 1983. Obtained through personal communication with J. Thiel, Radiation Control, Texas Department of Health, Austin, Texas.

---

* Memorandum undated, received by Texas Department of Health, Bureau of Radiation Control on September 25, 1984.

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

APPENDIX A

LIST OF COMPUTER DATA BASES AND YEARS SEARCHED

APPENDIX A

LIST OF COMPUTER DATA BASES AND YEARS SEARCHED

Agricola, 1970 - 1985

Aqualine, 1964 - 1985

Aquatic Science Abstracts, 1978 - 1985

Biosis Previews, 1969 - 1985

Chemical Abstracts Search, 1967 1985

DOE Energy Data Base, 1974 - 1985

Enviroline Data Base, 1970 - 1985

National Technical Information Service (NTIS), 1970 1985

Pollution Abstracts, 1970 - 1985

Water Resources Abstracts, 1958 1985

APPENDIX B

TELEPHONE SURVEY FORM AND LIST OF INTERVIEWEES

APPENDIX B

TELEPHONE SURVEY FORM

STUDY OF RADIOACTIVITY IN MUNICIPAL SLUDGE

TELEPHONE SURVEY

Date:

Person Contacted: Phone #:

Agency/Municipality:

For Battelle:

Summary:

Questions:

1. Does the "agency" have any reports or maintain a data base containing data on radioactivity levels in municipal sludges?

2. What cities or area of the country do the reports/data base cover?

3. Does the "agency" have data on wastewater or sludge concentrations of

   A. Radium?

   B. Gross Alpha?

   C. Gross Beta?

TELEPHONE SURVEY FORM (continued)

D. Individual radionuclides?

E. Gross Gamma?

4. What analytical methods were used? What are the limitations of the data?

5. Has the "agency" performed any special studies, for example, studies of uptake of radionuclides in crops grown in sludge amended soil?

6. Has the agency done any dose analysis for the sludge?

7. Is there any other information that you think would help with this work?

8. Can you send copies of any reports, data base information, special reports, etc? (Emphasize that our time frame is short so we'll need the reports/data soon.)

9. Do you know of any other agencies or other groups that have reported on or maintain a data base on radioactivity in municipal sludge?

Additional Notes:

LIST OF TELEPHONE SURVEY INTERVIEWEES

Federal Agencies and National Laboratories

Dr. Domy Adriano
Savannah River Ecology Laboratory
Aiken, SC
(803) 450-2475

Sam Aoki
Idaho Falls Nuclear Facility
U.S. Department of Energy (DOE)
Idaho Falls, ID
(208) 526-0583

Stan Arlt
Public Works Department
U.S. DOE
Richland, WA
(509) 575-2491

Bob Bastian
U.S. Environmental Protection Agency (EPA)
Washington, DC
(202) 382-7378

Tom Blumer
U.S. EPA
Region IX
San Francisco, CA
(415) 974-8338

Mr. Stanley Borys
Waste Treatment Department
Argonne National Laboratory
Chicago, IL
(312) 972-2000 (6677)

Dr. Rufus Chaney
U.S. Department of Agriculture
Agricultural Research Service
Beltsville, MD
(301) 344-3324

Ben Chen
U.S. EPA
Region IV
Atlanta, GA
(404) 881-4347

Frank Costello
U.S. Nuclear Regulatory Commission (NRC)
King of Prussia, PA
(215) 337-5000

Tony Drypolcher
Los Alamos National Laboratory
Los Alamos, NM
(505) 667-6378

Daniel Forger
U.S. EPA
Region II
New York, NY
(212) 264-0958

Don Galleogos
Los Alamos National Laboratory
Los Alamos, NM
(505) 667-5288

Norbert Golchert
Argonne National Laboratory
Chicago, IL
(312) 972-3912

LIST OF TELEPHONE SURVEY INTERVIEWEES (Continued)

Federal Agencies and National Laboratories (Continued)

Tom Gunderson
Environmental Surveillance
Los Alamos National Laboratory
Los Alamos, NM
(505) 667-5201

Tom Hakonson
Los Alamos National Laboratory
Los Alamos, NM
(505) 667-3331

Dick Hetherington
U.S. EPA
Region X
Seattle, WA
(206) 442-1941

Dick Lauch
U.S. EPA
Cincinnati, OH
(513) 569-7237

Russ Lease
Idaho Falls Nuclear Facility
U.S. DOE
Idaho Falls, ID
(208) 526-1668

Paul Lohouse
U.S. NRC
King of Prussia, PA
(215) 337-5246

Peter Ludzia
U.S. EPA
Region III
Philadelphia, PA
(215) 597-9226

J. R. Metzger
U.S. NRC
Washington, DC
(202) 492-7432

Donald M. Nelson
Industrial Hygiene
Argonne National Laboratory
Argonne, IL
(312) 972-4981

Mario Nunzio
U.S. EPA
Region VII
Kansas City, MO
(913) 236-2813

Tom Oakes
Oak Ridge National Laboratories
Oak Ridge, TN
(615) 576-8499

Max Richards
U.S. EPA
Region V
Chicago, IL
(312) 886-0122

Bob Schumaker
U.S. NRC
Office of Inspection and
Enforcement
Washington, DC
(202) 492-7432

Stanley Smith
U.S. EPA
Region VIII
Denver, CO
(303) 293-1563

LIST OF TELEPHONE SURVEY INTERVIEWEES (Continued)

Federal Agencies and National Laboratories (Continued)

Walt Smith
Argonne National Laboratory
Chicago, IL
(312) 972-3322

John Trax
Office of Drinking Water
U.S. EPA
Washington, DC
(202) 382-5526

LIST OF TELEPHONE SURVEY INTERVIEWEES (Continued)

State Agencies

Ken Arnold
Missouri Department of Natural Resources
Jefferson City, MO
(314) 751-3241

Stuart Blydenburgh
Office of Public Wastewater
Ohio Environmental Protection Agency
Columbus, OH
(614) 466-8945

Bruce Denny
Minnesota Radiation Control
Minneapolis, MN
(612) 623-5323

Brad Everson
Washington State Department of Energy
Olympia, WA
(206) 459-6291

Helen Fahy
Pennsylvania Department of Environmental Resources
P.O. Box 2063
Harrisburg, PA 17120
(717) 787-8184

Akos Fekete
New Jersey Department of Environmental Protection
Trenton, NJ
(609) 292-0424

Lavoy Haage
Waste Water Permits Branch
Iowa Department of Water, Air and Waste Management
Des Moines, IA
(515) 281-8975

Marty Hanrahan
Radiation Control Division
Colorado Department of Health
Denver, CO
(303) 320-8333

Al Hazale
Colorado Department of Health
Denver, CO
(303) 320-8333 (x6246)

Dave Heed
Illinois Department of Nuclear Safety
Springfield, IL
(217) 546-8100

Alice Hennigan
Radiation Control
Minnesota Radiation Agency
Minneapolis, MN
(612) 623-5323

George Johnson
Minnesota Water Quality Division
Minneapolis, MN
(612) 296-7169

LIST OF TELEPHONE SURVEY INTERVIEWEES (Continued)

State Agencies (Continued)

Francine Joyal
Bureau of Waste Management
Tallahassee, FL
(904) 488-0300

Jerry Lane
Division of Environmental Quality
Missouri Department of Natural Resources
Jefferson City, MO
(314) 751-3241

Terry R. Lash
Illinois Department of Nuclear Safety
Springfield, IL
(217) 546-8100

Gene Leiter
Montana Health Department
Helena, MT
(406) 444-2821

Paul Merges
New York Department of Environmental Conservation
Albany, NY
(518) 457-6603

Jim Milton
District Engineer for Richland Area
Washington Department of Ecology
Yacama, WA
(509) 456-2926

Ligia Mora
Solid Waste
Tallahassee, FL
(904)488-1344

Roy Ney
Water Supply
Iowa Department of Waste, Air, and Waste Management
Des Moines, IA
(515) 281-8945

Dan O'Neil
State of Michigan
Lansing, MI
(517) 373-8410

Ray Paris
Health Division
State of Oregon Radiation Control
Portland, OR
(503) 229-5797

Dick Pederson
Water Quality
Montana Health Department
Helena, MT
(406) 444-2406

Tom Potle
Bureau of Waste Water Management
Wisconsin Department of Natural Resources
Madison, WI
(608) 266-8343

Robert Quillen
Ohio Department of Health
Columbus, OH
(614) 466-3543

LIST OF TELEPHONE SURVEY INTERVIEWEES (Continued)

State Agencies (Continued)

Maggie Reiley
Radiation Protection Group
Pennsylvania Department of Environmental Resources
Harrisburg, PA
(717) 787-3479

Karim Rimawi
Director of Environmental Radiation
New York State Department of Health
Albany, NY
(518) 473-3613

Milton Rose
Texas Department of Water Resources
Austin, TX
(512) 463-8468

Roger Selberg
Illinois Environmental Protection Agency
Springfield, IL
(217) 782-9470

Dennis Sopich
Environmental Standards Division
Wisconsin Department of Natural Resources
Madison, WI
(608) 267-7570

Dr. Joseph Thiel
Texas Department of Health
1100 W. 49th Street
Austin, TX 78756-3189
(512) 835-7000

Mark Williams
Wisconsin Department of Natural Resources
Madison, WI
(608) 266-7278

LIST OF TELEPHONE SURVEY INTERVIEWEES (Continued)

Municipalities

Richard Borry
NY City Radiation Control
New York, NY
(212) 334-7774

Tom Bottenberg
Industrial Pretreatment
of Sewage Sludge
Portland, OR
(503) 796-7125

Jay Burnam
City of Portland Waste Water
Treatment
Portland, OR
(503) 285-0205

Dick Lanyon
Research & Development Section
Metropolitan Sanitary District of
Greater Chicago
Chicago, IL
(312) 751-5810

Peter MacNo
Seattle Metropolitan Water
Quality Department
Seattle, WA
(206) 447-6869

Stan Vendetti
Environmental Health Director
Benton/Franklin Health District
Richland, WA
(509) 943-2614

LIST OF TELEPHONE SURVEY INTERVIEWEES (Continued)

Sludge Distributors

Owen Anderson
Amchemco Marketing
Houston, TX
(713) 580-7803

Doug Boston
Kellogg Supply, Inc.
Wilmington, CA
(213) 830-2200

Joe Damas
Compgro
Los Angeles, CA
(415) 465-3700 (X120)

Mr. Don Gurda
Milorganite
735 North Water Street
Milwaukee, WI 53202
(414) 285-2266

Joe Harvath
Eko-Kompost, Inc.
Missoula, MT
(406) 721-1423

Henry Lebee
Compgro
Los Angeles, CA
(415) 465-3700

LIST OF TELEPHONE SURVEY INTERVIEWEES (Continued)

Other

L. L. Ames
Battelle Pacific
Northwest Division
Richland, WA
(509) 375-2121

Dr. Albert Bates
University of California
Riverside, CA
(714) 787-3654

Robert Buddermeier
Laurence Livermore Laboratory
Livermore, CA
(415) 422-1100

Terry Devine
Conference of Radiation
Control Directors
Frankfort, KY
(502) 227-4543

S. R. Joshi
National Water Research
Institute
Canada Centre for Inland Waters
Burlington, Ontario, Canada
(416) 336-4573

Dr. M. B. Kirkham
Evaportranspiration Lab
Dept. of Agronomy, Water Annex
Kansas State University
Manhattan, KS 66506
(913) 532-5731

Dr. Donald Lisk
Cornell University
Agricultural Experiment Station
Ithica, NY 14853
(607) 256-4538

Dr. Terry Logan
Ohio State University
Columbus, OH 43210
(614) 422-2001

C. W. Malody
Exxon Nuclear
Richland, WA
(509) 375-8100

P. M. McKee
Beak Consultants
6870 Goreway Dr
Missisaga, Ontario, Canada
L4V1P1
(416) 671-2600

Dr. Albert L. Page
Department of Soil Science and
Agricultural Engineering
University of California
Riverside, CA 92502
(714) 787-3654

Dr. Tony Peterlie
Ohio State University
Columbus, OH 43210
(614) 422-5520

LIST OF TELEPHONE SURVEY INTERVIEWEES (Continued)

Other, (Continued)

Howard M. Prichard
School of Public Health
Health Science Center
University of Texas
Houston, TX
(713) 792-4431

Ivan Schwabauer
Hygienics Laboratory
University of Iowa
Iowa City, IA
(515) 281-5371

Vernon Snoeyink
University of Illinois
Urbana, IL
(217) 333-4700

Vincent J. Sodd
Nuclear Medicine Lab
Cincinnati General Hospital
Cincinnati, OH
(502) 872-3100

M. D. Voss
Ames Laboratory
Ames, IA
(515) 294-2153

APPENDIX C

GAMMA EMISSIONS FROM SEWAGE SLUDGES

APPENDIX C

GAMMA EMISSIONS FROM SEWAGE SLUDGES

Table C-1. Results of a National Survey of Gamma Emissions from Sewage Sludges*

City	Gamma Radiation (cpm above background)
Baltimore, MD	5.4
Boston, MA	5.0
Dallas, TX	0.90
Denver, CO	7.2
Detroit, MI	3.9
Duluth, MN	1.1
Galveston, TX (Airport)	3.8
Galveston, TX (Main)	2.8
Houston, TX	5.9
Kalamazoo, MI	4.5
Knoxville, TN (Kuwahee)	5.7
Knoxville, TN (Fourth Creek)	4.2
Lexington, KY (Town Branch)	6.6
Lexington, KY (West Hickman Creek)	3.7
Los Angeles, CA	5.4
Memphis, TN (North)	2.9
Memphis, TN (T. E. Maxson)	5.4
Milwaukee, WI (South Shore)	4.8
Milwaukee, WI (Jones Island)	3.7
Philadelphia, PA (Northeast)	4.2
Philadelphia, PA (Southeast)	5.6
Phoenix, AZ	4.0
Portland, OR	4.5
Salt Lake City, UT	4.3
San Diego, CA	4.5

Table C-1 (continued)

City	Gamma Radiation (cpm above background)
San Francisco, CA (Richmond-Sunset)	1.7
San Francisco, CA (North Point)	1.8
Seattle, WA	2.6
Syracuse, NY	3.9
Toledo, OH	2.3
Groton, NY	3.2

* Source: Mumma et al., 1984

Table C-2. Results of a Survey of Central New York State Sewage Sludges*

City	Gamma Radiation (cpm above background)
Batavia	2.5
Bath	1.0
Canandaigua	4.4
Canisteo	2.7
Clyde	2.7
Corning	6.5
Geneseo	2.9
Geneva	9.7
Henrietta	3.5
Holcomb	2.8
Honeoye Falls	3.0
Hornell	3.0
Leroy	6.5
Lyons	1.3
Macedon	0.15
Manchester	3.0
Medina	3.8
Montour Falls	4.9
Mt. Morris	5.1
Newark	2.2
Palmyra	2.3
Phelps	2.0
Scottsville	2.3
Sodus	0.54

* Source: Mumma et al., 1983

APPENDIX D

BATTELLE STAFF CONTRIBUTING TO PREPARATION OF DOCUMENT

APPENDIX D

This document was prepared for the Criteria and Standards Division of the U.S. Environmental Protection Agency under Contract No. 68-01-6986 to Battelle Columbus Division, Columbus, Ohio. The following Battelle staff contributed to the prepartion of this document:

Susan E. Brauning - Work Assignment Leader
Peter J. Ferron
Scott J. Maris
John T. McGinnis
Barney W. Cornaby
Andrea C. Hall
Kenneth M. Duke