Protecting Our Water Environment

Metropolitan Water Reclamation District of Greater Chicago

RESEARCH AND DEVELOPMENT DEPARTMENT

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RADIOACTIVITY IN BIOSOLIDS-AMENDED SOIL AND UPTAKE BY CORN

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DISCLAIMER

Mention of proprietary equipment and chemicals in this report does not constitute endorsement by the Metropolitan Water Reclamation District of Greater Chicago

SUMMARY

Wastewater treatment at publicly owned treatment works (POTWs) produces a considerable amount of biosolids. These biosolids are rich in plant nutrients, and may be used for beneficial purposes such as application to agriculture fields and disturbed lands. The Metropolitan Water Reclamation District of Greater Chicago (District) is one of the largest municipal wastewater treatment agencies in the nation, and it produces about 190,000 dry tons of biosolids each year. The District monitors the quality of biosolids produced, and is committed to using it beneficially. One beneficial use is fertilizing or reclaiming disturbed lands (mine soil) for crop production. The District has made long-term applications of biosolids to mine soil at the Corn Fertility Experimental Plots in Fulton County, Illinois. Biosolids were applied annually at the rates of 16.8, 33.6, and 67.2 Mg ha^{-1} to replicated plots (4 times) from 1973 to 2000 along with commercial fertilizer, which was applied annually to control plots. The purpose of this study was to evaluate the effect of long-term applications of biosolids on the radioactivity concentration in biosolids-amended soil, and the uptake of radioactivity into corn grain and stover harvested from these plots. Radiological analyses show nearly the same consistent concentration of radioactivity in soil from the biosolids-amended plots as that found in the soil from control plots receiving commercial fertilizer. The same trend was observed for corn grain and stover harvested from these plots. Of the twenty-seven radionuclides studied, only potassium-40 was found in corn grain and stover, and a negligible amount of radium-226 was observed in corn grain. No other radionuclide was found in corn grain and stover. The results of this study show that long-term annual applications of biosolids did not increase the radioactivity in amended soil or result in increased uptake by corn. The low or non-detectable levels of radioactivity observed in soil and corn indicate that the District's land applied biosolids do not pose a threat to human and animal health or the environment from a radioactivity standpoint.

INTRODUCTION

The Metropolitan Water Reclamation District of Greater Chicago is located within the boundaries of Cook County, Illinois. It is one of the largest municipal wastewater treatment agencies in the nation and operates seven water reclamation plants (WRPs). It serves an area of 0.226 Mha including the City of Chicago and 125 suburban communities with a combined population of 5.1 million people. In addition, a waste load equivalent of 4.9 million people is contributed within the District's service area by industrial and commercial sources. On the average, the District treats 1,500 million gallons per day (MGD) of wastewater at its seven WRPs.

Radioactivity in the sewerage system may enter from a variety of sources including industries, hospitals, research organizations, and licensed radioactive material users (ISCORS 2003). Naturally occurring radionuclides and those in atmospheric fallout also enter the sanitary sewerage system from groundwater and through stormwater runoff. The purpose of wastewater treatment is to remove pollutants from raw sewage to ensure adequate effluent quality and reduce its adverse impact on the receiving surface waters. The low concentrations of radioactive materials from natural and man-made sources discharged to the sanitary sewerage system may become concentrated in biosolids during wastewater treatment and sludge processing. The District produces approximately 190,000 dry tons of biosolids each year. These biosolids are rich in plant nutrients. The District looks at beneficial ways to use these biosolids. The beneficial uses include application to parks, golf courses and athletic fields; agricultural lands to fertilize crops; strip-mined or other disturbed soils for reclamation, which may result in crop production; and landfills as a soil conditioner or soil substitute in the final cover.

Radioactivity in soil is a potential source of contamination for food and forage. Radionuclides may accumulate in plants either from atmospheric deposition on the leaves and shoots or through uptake by the roots from the soil. Radioactivity contaminated plants pose a risk to human health either through direct ingestion of crops, or indirectly from the ingestion of milk from animals raised on contaminated forage. The District owns a large tract of land in Fulton County, Illinois (6,122 ha). This site is used to recycle biosolids for the purpose of reclaiming mine soil and to fertilize agricultural crops. The District has made long-term applications of biosolids on mine soil at the Corn Fertility Experimental Plots in Fulton County, Illinois. The biosolids were applied annually at the rates of 16.8, 33.6, and 67.2 Mg ha^{-1} to replicated plots (4 times) from 1973 to 2000 along with commercial fertilizer, which was applied annually to control plots. Soils, corn grain, and corn stover from this site were sampled in 2000 for this study. The purpose of this study was to evaluate the effects of long-term high rate applications of biosolids on soil radioactivity and the uptake of radioactivity by corn harvested from biosolids-amended soils.

MATERIALS AND METHODS

Plot Description and Sample Collection

The Corn Fertility Experiment is a random complete block design with four treatment plots contained in each of four replicate blocks. Each plot is 12.2 m long and 4.6 m wide. The treatments consist of control (no biosolids, fertilized with 336-224-112 kg ha^{-1} of N-P-K annually), and biosolids-amended plots receiving annual application rates of 16.8 Mg ha^{-1} (quarter-maximum), 33.6 Mg ha^{-1} (half-maximum), and 67.2 Mg ha^{-1} (maximum). The biosolids plots also receive 112 kg ha^{-1} of potassium fertilizer annually. The cumulative biosolids loadings that were received by the quarter-maximum, half-maximum, and maximum amended plots from 1973 through 2000 were 455, 909, and 1,817 Mg ha^{-1}, respectively.

Composite soil samples were collected in each plot in 2000 at the 0 to 15 cm depth. Corn grain and stover were harvested from 10.7 m strips within the middle two rows of each plot. Representative samples of each were collected from the harvested matter in each plot.

Sample Preparation

Corn grain and stover samples were washed and dried at 65° C for 48 hours. Samples were then ground in a Wiley mill and sieved through a 20-mesh stainless steel screen. Soil samples were air-dried and ground to pass a 2-mm sieve (Pietz et al. 1983).

Gross Alpha and Beta Radioactivity

Gross alpha and beta radioactivity concentrations in the samples were determined using Standards Methods for the Examination of Water and Wastewater procedure (Standard Methods 1998). A thoroughly mixed sample of soil, corn grain, or corn stover was transferred to a tared evaporating dish. The sample was dried in an oven to constant weight at 103° C. The difference in weight of the dried sample over the empty dish represents the total sample dry weight. The dried sample was then transferred to a cool muffle furnace. The temperature of the furnace was raised gradually over a period of three hours to 550° C, and the sample was incinerated overnight at this temperature. The residue in the dish represents the fixed solids. The fixed solids were ground to a fine powder. A weighed portion of the fine powder (80 to 100 mg) was transferred to a tared stainless planchet. The residue was distributed to a uniform thickness and spread with a few drops of 0.5% (w v^{-1}) Lucite solution in acetone. It was then dried to constant weight at 103° C and counted for gross alpha and gross beta radioactivity on Tennelec LB5100 Gas Proportional Counter. A National Institute of Standards and Technology (NIST) traceable thorium-230 standard for gross alpha and cesium-137 standard for gross beta radioactivity were used for efficiency calibration of gas proportional counters.

Gamma Radioactivity

The dried soil, corn grain, or corn stover sample was packed in a tared 85-g canister, and then weighed and sealed with a vinyl electrical tape to avoid the loss of the gaseous progeny of uranium and thorium. The sample was stored for at least 30 days for radon-radium to reach equilibrium before counting. The sample was analyzed by gamma spectroscopy. A list of radionuclides monitored is given in Table 1.

TABLE 1: LIST OF RADIONUCLIDES MONITORED.

Beryllium-7	Sodium-22	Potassium-40	Manganese-54
Cobalt-57	Cobalt-60	Zinc-65	Niobium-94
Ruthenium-106	Silver-108m	Silver-110m	Antimony-125
Cesium-134	Cesium-137	Cerium-144	Europium-152
Gadolinium-153	Europium-154	Europium-155	Bismuth-207
Bismuth-212	Lead-212	Bismuth-214	Lead-214
Radium-226	Actinium-228	Protactinium-231	

The gamma spectroscopy system was equipped with a p-type coaxial high purity germanium detector with a relative efficiency of 25% and a resolution of 1.8 keV full width at half maximum at 1,332.5 keV gamma transition of Co-60. The spectroscopy system was also equipped with a Ginie-2000 spectroscopy software package from Canberra Industries. The detector was shielded by a 10-cm thick low background virgin lead cylindrical shield with a fixed bottom, and a moving cover and graded lining of 0.5 mm cadmium and 1.6 mm copper to reduce gamma ray background.

The gamma spectrometer was calibrated with a NIST traceable 9 radionuclide mixed gamma standard from North American Scientific, Inc., density 1.07 g cm^{-3}, and packed in a 85-g canister with an energy range of 88 keV to 1,836 keV. The energy and efficiency calibration of the system was verified before each use. The radium-226 radioactivity concentration was evaluated from 186.2 keV photopeak, cesium-137 from 661.6 keV photopeak, bismuth-212 from 727.2 keV photopeak, lead-212 from 238.6 and 300.1 keV photopeaks, bismuth-214 from 609.3 and 1,120.3 keV photopeaks, and lead-214 from 295.2 and 351.9 keV photopeaks. The activity of potassium-40 was derived from 1,460.8 keV photopeak, and actinium-228 from 338.3, 911.6, and 969.1 keV photopeaks.

Statistical Analysis

Statistical analysis was performed using the analysis of variance (ANOVA) to test for the significant differences (Walpole and Myers 1989).

RESULTS AND DISCUSSION

The gross alpha and gross beta radioactivity concentration in soil, corn stover, and corn grain harvested from the Corn Fertility Experiment in 2000 is presented in Table 2. The data represents the means of four-sample analyses for each treatment.

The gross alpha radioactivity concentration in soil from control plots was 340 Bq kg^{-1} dry weight (dw), and 293, 318, and 352 Bq kg^{-1} dw soil from the quarter, half, and maximum biosolids-amended treatment plots, respectively. The gross beta radioactivity concentration in soil from control plots was 1,062 Bq kg^{-1} dw, and 1,092, 1,090, and 1,066 Bq kg^{-1} dw soil from the quarter, half, and maximum biosolids-amended treatment plots, respectively.

The gross alpha radioactivity in corn stover harvested from the control plots was less than 7 Bq kg^{-1} dw, and in the quarter, half, and maximum treatment plots it was 18, 15, and less than 8 Bq kg^{-1} dw, respectively. The gross alpha radioactivity in corn grain was less than 2 Bq kg^{-1} dw in all the samples. The gross beta radioactivity in corn stover harvested from control plots was 471 Bq kg^{-1}, and in the quarter, half, and maximum treatment plots it was 422, 471, and 505 Bq kg^{-1} dw, respectively. The gross beta radioactivity in corn grain harvested from control plots was 143 Bq kg^{-1} dw, and in the quarter, half, and maximum treatment plots it was 138, 163, and 144 Bq kg^{-1} dw, respectively.

TABLE 2: CONCENTRATIONS OF GROSS ALPHA AND GROSS BETA RADIOACTIVITY IN SOIL, CORN STOVER, AND CORN GRAIN FROM BIOSOLIDS-AMENDED MINE SPOIL.

	Gross Alpha Radioactivity (Bq kg^{-1} dw)			Gross Beta Radioactivity (Bq kg^{-1} dw)		
Treatment[a]	Soil	Stover	Grain	Soil	Stover	Grain
Control	340±64[b]	<7	<2	1,062±32	471±35	143±19
Quarter	293±38	18±7	<2	1,092±90	422±54	138±24
Half	318±42	15±3	<2	1,090±36	471±92	163±18
Maximum	352±75	<8	<2	1,066±22	505±89	144±32
ANOVA	NS[c]	*	NS	NS	NS	NS

[a] Plots in the control treatment received 336-224-112 kg ha^{-1} of N-P-K annually, and plots in the quarter, half, and maximum treatments received 455, 909, and 1,817 Mg ha^{-1} of biosolids from 1973 through 2000 and 112 kg ha^{-1} K annually.

[b] Mean ± standard deviation.

[c] Not significantly different.

*Significant at the 0.05 level of probability.

The gross alpha radioactivity concentrations in soil and corn stover and grain did not reflect the different cumulative biosolids applications that were made to the quarter, half, and maximum treatment plots. The ANOVA's for gross alpha radioactivity concentrations in soil and corn grain (Table 2) show that there were no significant differences at 5% level between the biosolids-amended and control plots. The ANOVA's also show that the gross beta radioactivity concentrations in soil and corn stover and grain were not significantly different between the biosolids-amended and control plots (Table 2). This indicates that there was no increase in the uptake of gross beta radioactivity by corn grain from biosolids-amended soil.

The concentrations of gamma-emitting radionuclides in biosolids-amended soil and corn stover and grain harvested from these plots were determined. Of the 27 radionuclides monitored, 8 were detected at measurable levels in the soil of the control and biosolids-amended plots as shown in Table 3. Of those, 7 radionuclides are of natural origin, and one, cesium-137, is a man-made radionuclide. Only potassium-40 was detected at measurable concentrations in stover, whereas only potassium-40 and radium-226 were detected at measurable levels in grain. The concentration of all other radionuclides monitored was below the detection limit.

The data in Table 3 show that the concentrations of gamma-emitting radionuclides in soil from the control plots were similar to the radioactivity concentrations in biosolids-amended plots, except for potassium-40 and cesium-137 where significant differences were observed between the treatments. Potassium-40 was lower in the soil of the maximum treatment, and cesium-137 was slightly higher in the maximum treatment as compared to the other treatments. The concentrations of potassium-40 in the stover from biosolids-amended plots were not significantly different than the concentrations in stover from the control plots. In corn grain the concentrations of potassium-40 were not significantly different between the control and biosolids-amended plots. The ANOVA shows that the concentrations of radium-226 in corn grain varied significantly between the treatments because of the variability in concentrations (Table 3). The concentrations of radium-226 in the biosolids plots were similar to those observed in the control plots.

Small amounts of bismuth-212, lead-212, bismuth-214, lead-214, actinium-228, and cesium-137 were found in soil from control and biosolids-amended plots, but these radionuclides were not detected in stover and grain from any of the plots. The data indicate that there was no uptake of gamma emitting radionuclides by corn stover and grain as a result of biosolids applications to land above the normal background level, even at cumulative loading rates of as high as 1,817 Mg ha^{-1}. The radiological analyses data show that long-term biosolids applications to the Corn Fertility Experimental Plots on mine soil did not increase the radioactivity concentration in biosolids-amended soil or result in the uptake of radioactivity by corn.

TABLE 3: CONCENTRATIONS OF GAMMA-EMITTING RADIONUCLIDES IN SOIL AND CORN STOVER AND GRAIN FROM BIOSOLIDS-AMENDED MINE SPOIL.

	Treatment[a]				
Radionuclides	Radioactivity (Bq kg^{-1} dw)				
	Control	Quarter	Half	Maximum	ANOVA
			Soil		
Potassium-40	634±36[b]	640±29	634±13	585±19	*
Radium-226	120±15	119±11	123±12	131±9	NS[d]
Bismuth-212	27±4	27±4	26±0	24±2	NS
Lead-212	39±2	39±2	38±2	38±2	NS
Bismuth-214	40±4	41±4	40±2	39±2	NS
Lead-214	44±3	46±5	45±2	46±2	NS
Actinium-228	41±4	39±2	39±2	38±2	NS
Cesium-137	2±1	2±1	2±2	4±1	*
			Corn Stover		
Potassium-40	545±45	485±115	496±105	511±78	NS
Radium-226	nd[c]	nd	nd	nd	-
Bismuth-212	nd	nd	nd	nd	-
Lead-212	nd	nd	nd	nd	-
Bismuth-214	nd	nd	nd	nd	-
Lead-214	nd	nd	nd	nd	-
Actinium-228	nd	nd	nd	nd	-
Cesium-137	nd	nd	nd	nd	-
			Corn Grain		
Potassium-40	115±11	113±9	117±6	113±5	NS
Radium-226	24±2	28±2	23±2	27±2	*
Bismuth-212	nd	nd	nd	nd	-
Lead-212	nd	nd	nd	nd	-
Bismuth-214	nd	nd	nd	nd	-
Lead-214	nd	nd	nd	nd	-
Actinium-228	nd	nd	nd	nd	-
Cesium-137	nd	nd	nd	nd	-

[a] Plots in the control treatment received 336-224-112 N-P-K kg ha^{-1} annually, and plots in the quarter, half, and maximum treatments received 455, 909, and 1,817 Mg ha^{-1} of biosolids from 1973 through 2000 and 112 kg ha^{-1} K annually.

[b] Mean ± standard deviation.

[c] Not detected.

[d] Not significantly different.

* Significant at the 0.05 level of probability.

CONCLUSIONS

Long-term biosolids applications to disturbed lands did not significantly effect the concentrations of radionuclides in the biosolids-amended soil or in corn tissues. Potassium-40 was the only radionuclide detected in both corn stover and grain, and radium-226 was detected only in grain. As expected, the concentrations of potassium-40 were lower in corn grain as compared to soil and corn stover. Since there were no significant differences in the concentrations of potassium-40 in corn tissues from the control and biosolids-amended soils, the radium-226 concentrations in the biosolids plots were similar to those in the control plots, and no other radionuclides were even detected in corn tissues in this study, it is concluded that the consumption of corn produced on the biosolids-amended soil by livestock is not likely to increase the risk of exposure from radioactivity to humans and animals or the environment.

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