

IMPACT OF LONG-TERM LAND APPLICATION OF BIOSOLIDS ON GROUNDWATER QUALITY AND SURFACE SOILS*[†]

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ABSTRACT

A study was conducted to evaluate the long-term land application of sewage sludge and its potential impact on groundwater quality and surface soils. For this study, an existing site that has been in operation for more than fifteen years was selected for sampling and analyses. From this site, sludge applied soil samples, background soil samples, and groundwater monitoring samples were obtained. The samples were analyzed for the following: pH, conductivity, total solids, fecal coliform, fecal streptococci, nitrate nitrogen, ammonia nitrogen, TKN, arsenic, cadmium, chromium, copper, nickel, lead, and zinc. The results of this study indicate that groundwater at this sludge application site was not contaminated with heavy metals or pathogens. However, in some instances the groundwater nitrate nitrogen concentrations were slightly higher than background levels due to excessive nutrient loadings. This problem can be alleviated by applying sludge at agronomic rates so that no excess nitrogen is available for leaching. The bacteriological soil data indicated that the levels of fecal coliform and fecal streptococci were similar to background levels with no evidence of contamination.

*Funding for this work was provided, in part, by U.S. EPA and the Missouri Department of Natural Resources, and, in part, by the University of Missouri, Columbia, Missouri.

[†]The views/opinions expressed in this article are those of the authors and should not be construed as opinions of the United States Environmental Protection Agency.

INTRODUCTION

One of the most common disposal practices for sewage sludge is land application. Approximately 33 percent of the sewage sludge used or disposed annually in the United States is applied to land [1]. In recent years, more and more wastewater treatment plants are land applying sewage sludge instead of disposing of it by land filling, ocean dumping, or incineration. Sewage sludge contains nutrients and organic matter, including nitrogen, phosphorus and lesser quantities of potassium, as well as other essential nutrients beneficial for growing crops [2]. Sewage sludge contains \$50 to \$60 of nutrient and soil conditioning value per dry ton [3]. Many researchers have reported that sludge-treated soil produced crop yields equivalent to or higher than those when commercial fertilizers alone were applied [4-6]. The U.S. Environmental Protection Agency (EPA) promulgated regulations (40 CFR Part 503) to protect public health and the environment from any reasonably anticipated adverse effects of certain pollutants that may be present in sewage sludge [1]. The regulations establish standards for the final use and disposal of sewage sludge. In developing these use and disposal regulations, EPA evaluated the potential risks to public health or the environment from individual pollutants present in sewage sludge. EPA risk analyses evaluated fourteen pathways of potential exposure to pollutants in sewage sludge.

The major concerns involved in land application of sewage sludge include possible surface and groundwater contamination by excessive applications of nitrogen, phosphorus, and organic materials; accumulations of heavy metals and trace organic chemicals; pathogenic microorganisms; and food-chain contamination with toxic elements [7].

The present study evaluates the long-term land application of sewage sludge and its potential impact on groundwater quality and surface soils. A study was conducted by sampling and evaluating data from a successfully operating long-term sludge application site. The purpose of this study was to evaluate the actual effects of long-term sludge application on groundwater quality, soil characteristics, and any other human health or environmental concerns. These studies were done to help interpret the data generated from the field studies.

BACKGROUND

Sludge Application Rate

Application rates of sewage sludge to farmland vary according to the nutrient requirements of the crops, existing soil characteristics (e.g., drainage, nutrient levels, heavy metal content, etc.), climate, and the characteristics of the sludge. In all cases, the application rate should be such that (a) crop production and

quality are not decreased; (b) the soil does not accumulate excessive organic matter and heavy metals; (c) nutrients and excessive salts do not leach into the surface or groundwater [8]. For the best management of sludge application, the nutrient supply should be adequate for the crop's needs and the metal loading should not exceed the ceiling concentration and the cumulative pollutant rate [1].

Nutrients Management

Sewage sludge contains considerable quantities of nitrogen and phosphorus but low quantities of potassium as a source of plant fertilizer. Generally, the nutrient requirement for sludge application is based on the crops species and the soil properties [2, 7]. Not all of the nitrogen in sewage sludge is immediately available to plants as some is present as organic nitrogen. Organic nitrogen must be decomposed into mineral or inorganic forms of nitrogen like ammonia-nitrogen and nitrate nitrogen before plants can use it. Accordingly, the Plant Available Nitrogen (PAN) depends on the microbial breakdown of the applied organic materials in soils, as does the available nitrogen present in other organic matter in the soil [7]. For adequate utilization of sewage sludge, the agronomic rate should be determined prior to land application of sludge.

In the EPA 40 CFR 503, agronomic rate is defined as the whole sludge application rate designed: 1) to provide the amount of nitrogen needed by the crops or vegetation grown on the land, and 2) to minimize the amount of nitrogen in the water that passes below the root zone of the crops or vegetation grown on the land to the groundwater [1].

To determine this rate, the nitrogen needs of the crops, the Plant Available Nitrogen (PAN) in the sewage sludge, the soil conditions at the site, and the geology of the site have to be known.

Nitrates in Groundwater

The potential hazard most frequently associated with nutrients in sewage sludge is the mismanagement that can cause pollution of water resources, especially with nitrate-nitrogen contamination of groundwater [2, 9]. The importance of groundwater is that it is the main source of drinking water for about half of the U.S. population and for about 85 percent of the rural population [10]. When nitrate nitrogen is ingested in high enough amounts by human beings and animals, potential adverse health effects may occur. These effects are reported to include *methemoglobinemia*, *cancer*, and possibly others [9, 10].

Being anionic and largely unreactive with soil particles, nitrate in solution is highly mobile in the soil until it is immobilized by microorganisms or assimilated by plants. Thus, it moves readily with soil water and may be leached out of the rooting zone of the crop [11]. Hinesly et al. reported that the application of 1,764 kg ammonia-nitrogen/ha in liquid sludge resulted in nitrate-nitrogen

concentrations in soil leachates of > 100 mg/L [12]. These excessive nitrate-nitrogen levels in the soil solution are to be anticipated because plant available nitrogen added was ten to twenty times that required by the crop grown. This study concluded that a sludge application rate of 15 metric tons/ha would minimize the amounts of nitrate leached into groundwater. Soon et al. concluded from a study designed to compare nitrate levels in soils treated with NH_4NO_3 and digested sludge that minimal nitrate pollution of groundwater would occur if the amount of nitrogen applied in sludge was consistent with the nitrogen requirement of the crop grown [13]. Therefore, Stewart et al. suggested that the sludge application rate should be based on the need for a specific crop because additional amounts of sludge did not increase crop yields but increased the risk of nitrate-nitrogen leaching into the groundwater [14].

The important factors that affect nitrate nitrogen leaching into groundwater include seasons, irrigation rate, rainfall, soil types, crops utilization rate and elevation of groundwater table, and sludge application rate [15, 16]. Nitrate nitrogen leaching from the root zone to the aquifers mainly takes place in the autumn till spring period when precipitation exceeds evaporation. The leaching of nitrate nitrogen normally occurs at the end of the growing season when the crops are not growing, so that residual nitrate nitrogen in the top soil will be readily leaching into groundwater. Therefore, residual nitrate-nitrogen in the root zone should be as low as possible by the end of the growing season [16].

Heavy Metals in Sludge-Amended Soil

In addition to nutrients, sewage sludge contains a number of potentially harmful constituents such as heavy metals. The presence of heavy metals in applied sludge creates the potential of soil and groundwater contamination. The extent of a heavy metal problem depends on the composition of sludge, sludge application rate, and soil properties [17].

Sewage sludge contains a wide range of heavy metal concentrations. Ten metals that are of most concern to plant, animal, and human health are: Arsenic, Cadmium, Chromium, Copper, Lead, Mercury, Molybdenum, Nickel, Selenium, and Zinc. These metals are also included in the pollutant limits set by EPA (40 CFR 503). Of these, Cadmium is regarded as potentially the most hazardous metal element when sludge is applied on land [18]. In most cases the metal concentrations in sewage sludge are below currently acceptable limits for land application.

Movement of metals from sludge-amended soil is related to soil pH [18, 19]. Generally, maintaining a soil pH of 6.5 or higher immobilizes most cationic heavy metals in the soils [7, 17]. Boswell reported the movement of Zn to a depth of 30 cm and Cd, Cr, Cu to 15 cm after sludge application of 168 metric tons/ac (415 metric tons/ha) to the soil [20]. The soil pH dropped from 6.2 to 5.0 during the 2-year study. Hinesly et al. reported the movement of Zn, Cd, Cr, Cu, and Ni

to a depth of 30 to 45 cm in an agricultural soil after application of 136 metric tons/ac (336 metric tons/ha) sludge over a four-year period; the soil pH dropping from 5.6 to 4.9 at the end of two years [12]. These lowered pH's could have increased the solubility of the metals and contributed to heavy metal movement.

Yangming and Corey studied the vertical and horizontal redistribution of Cd, Cu, and Zn in a silt loam plot where sludge had been applied in 1978 [21]. The sludge was incorporated by plowing and disking into the upper 20 cm layer, and crops (barley and sorghum) were grown for the next ten years. The soil sampling in 1989 revealed that in the upper layer (0 to 20 cm) the distribution of sludge-borne Cd, Cu, and Zn were 66.7, 67.5, and 68 percent, respectively. Some of the heavy metals (15.5 to 21%) moved outside the plot area, while a small amount (11.6 to 15.2%) was found in the deeper layer (20 to 30 cm). No significant accumulations of these metals were found below a depth of 30 cm.

In a controlled column study, Chang et al. evaluated the movement of Cd, Cu, Ni, and Zn, and indicated that no metals moved below the sludge-soil layer after twenty-five months [22]. Dowdy and Volk indicated that sludge-borne metals (Cd, Cu, and Zn) remained in the top 1.0 m of soil after fourteen years of massive sludge additions (765 metric tons/ha cumulative sludge application, on a dry weight basis) [23]. They also presented a rather extensive review of available research data for leaching of sludge-borne metals in sludge-treated soil. They found very little evidence of trace metals movement beyond the incorporated zone.

Fresquez et al. reported that the application of sewage sludge to a degraded semiarid grassland soil insignificantly increased soil N, P, and K, but without increasing heavy metal elements, such as Cadmium and Lead [24]. Robertson et al. indicated that extractable Copper remained in the top 30 cm and Zinc in the top 90 cm of the profile after application of 355 metric tons/ha on a dry weight basis over a six-year period [25]. The levels of Cd, Cr, and Ni in the sludge-treated soil were too low to be accurately determined. Martens et al. reported that the maximum Diethylenetriaminepentaacetic acid (DTPA) extractable metal levels in the soils were 0.6 mg Cd/kg, 150 mg Cu/kg, 4.0 mg Ni/kg, and 75 mg Zn/kg after 210 metric tons/ha sludge (dry solids) were applied [26]. The highest metal loading rates were 4.5 kg Cd/ha, 760 kg Cu/ha, 43 kg Ni/ha, and 620 kg Zn/ha. Phytotoxicity did not occur in the corn grown on these diverse soils, even where Cu and Zn were applied in excess of USEPA guidelines.

Accordingly, the movement of heavy metals in the sludge-amended soil can be reduced when neutral to basic soil pH is maintained in soil profiles [27]. Under these conditions most of the metals accumulated in the surface sludge-soil layer even after repeated sludge land applications. Therefore, groundwater contamination is unlikely when sewage sludge is applied to a well designed field under controlled loading conditions.

MATERIALS AND METHODS

For this study, an existing sludge application site which has been active for more than fifteen years was selected. This site was visited to obtain: 1) soil samples where sludge has been applied; 2) background soil samples; and 3) groundwater monitoring samples. In addition, the treatment plant provided several years of sludge, soil, and groundwater monitoring data for analysis and evaluation.

Wastewater Treatment Facility

Primary treatment at this treatment facility consists of a bar screen, grit chamber, and a primary clarifier. Secondary treatment consists of two activated sludge aeration basins followed by secondary clarifiers. The primary sludge is thickened in a gravity sludge thickener. The waste activated sludge is thickened by centrifugation, and then combined with thickened primary sludge and introduced to a two-sludge anaerobic digester system which contains three primary digesters and one secondary digester. The digested sludge is stored in a sludge storage lagoon. The stabilized sludge is then injected to a field nearby or off-site to privately owned farmland. The on-site injection program was started in 1984, and the total injectable area is 138.1 acres (55.9 ha). Since then, the average on-site sludge application rate has been 6.0 dry tons per acre per year (13.5 metric tons per ha per year) (Table 1) and the average off-site sludge application rate has been 2.6 dry tons per acre per year (6.5 metric tons per ha per year). The lifetime cumulative application rate was 36.4 dry tons per acre (81.1 metric tons/ha). Four groundwater observation wells are located at the injection site to monitor groundwater quality. Quarterly water quality data from the groundwater wells were available from 1987-1993.

Sources of Samples

A set of samples were taken from the sewage sludge injection site (Figure 1). The sludge-amended soil samples were collected from areas within each site that had sludge applied recently. Samples were taken from the surface layers (0 to 5 cm), as well as deeper layers (20 to 25 cm). The background soil samples (untreated soil) were also collected to compare with the sludge-amended soils. Groundwater samples were collected from monitoring wells.

Sample Handling

Sludge-amended soil samples were collected and placed in zippered plastic bags. Sterilized plastic containers with sealable tight fitting covers were used for groundwater and sludge samples. The sampling procedures were according to EPA publication, "POTW Sludge Sampling and Analysis Guidance Document" [25]. Samples were analyzed at once upon arrival at the laboratory for the

Table 1. Sludge Application Rate

	Volume Injected (m ³)	Solids (%)	Area Injected (ha)	Sludge ^a Injected (mt)	Annual ^a Application Rate (mt/ha)
ON-SITE					
1984	6,139	3.90	16.60	239.17	14.41
1985	18,031	3.21	21.89	603.63	27.57
1986	11,815	2.35	25.16	303.66	12.06
1987	22,963	2.40	33.20	519.48	15.69
1988	35,444	3.40	55.87	1,107.26	19.82
1989	10,626	3.50	50.36	373.88	7.42
1990	13,325	3.90	33.31	521.10	15.69
1991	5,587	3.30	17.05	185.16	10.76
1992	11,060	4.70	50.36	520.19	10.31
1993	4,681	5.10	50.36	238.50	4.71
Average	13,967	3.58	35.42	461.20	13.84
OFF-SITE					
1987	16,380	5.90	151.18	1,068.39	7.06
1988	7,752	3.80	58.63	293.13	5.00
1989	30,015	4.50	266.77	1,352.81	5.07
1990	19,300	5.20	155.53	1,005.81	6.50
1991	848	3.40	6.07	29.21	4.71
1992	15,814	5.10	125.34	813.58	6.50
1993	14,466	4.90	122.75	704.62	5.83
Average	13,691	4.61	108.01	663.03	6.58

^aDry Weight Basis

following: pH, conductivity, total solids, fecal coliform, fecal streptococci, nitrate nitrogen, ammonia nitrogen, arsenic, cadmium, chromium, copper, nickel, lead, and zinc. Samples were analyzed by the procedures listed in *Standard Methods for the Examination of Water and Wastewater* [29] and according to EPA sludge sampling and analysis guidance document [28].

RESULTS AND DISCUSSION

The data presented hereinafter includes monitoring data supplied by the wastewater treatment plant and data generated from sampling and analyses of soil obtained from site visits to the land application fields.

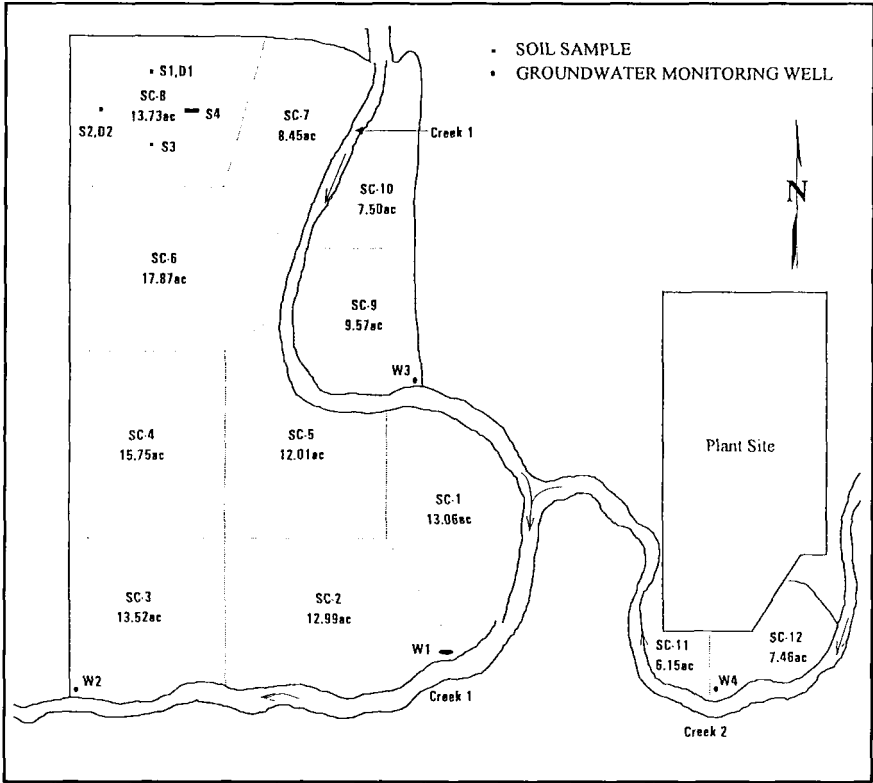


Figure 1. Sludge injection site.

Nitrogen Loadings

The nitrogen requirement of the crops was met by that supplied by the soil and that added in the form of thickened liquid sludge. The amount of nitrogen supplied by the soil was estimated from the organic content of the soil and the cation exchange capacity. Blanchar et al. reported that the soil present at the sludge disposal sites of the plant contained an average of 36 kg N/ha/yr [30].

The injection sites were cultivated with three different types of crops. The main crop was corn. Corn was grown every year on some sites, while wheat and soybeans were rotated every other year on other sites. For the analysis of N uptake by plants, the following criteria were assumed based on the recommendations by USEPA [31]:

- For corn yield of 11.8-13.4 metric tons/ha, N uptake = 258 kg/ha.
- For wheat yield of > 4.6 mt/ha, N uptake = 100 kg/ha.
- For soybeans yield of > 4.6 mt/ha, N uptake = 336 kg/ha.

Nitrogen analysis of the sludge is presented in Table 2. Total nitrogen, ammonia nitrogen, organic nitrogen, and nitrate nitrogen were measured on the basis of dry sludge. The ammonia nitrogen concentrations in the digested sludge varied from 19,760 mg/kg to 53,300 mg/kg. The maximum concentration of organic nitrogen in the sludge during the entire sludge injection operation was 55,400 mg/kg, whereas the minimum concentration was 15,750 mg/kg. Nitrate concentrations in the sludge were very low, averaging about 100 mg/kg. Plant Available Nitrogen (PAN) was calculated using guidelines of the Missouri Department of Natural Resources [32].

Table 3 shows the nitrogen loading rates of the digested sludge applied to the fields for all years of the sludge injection operation. The PAN of the sludge applied varied from year to year with a low value of 117 kg/ha in 1993 to a high of 1148 kg/ha in 1985. Table 4 shows the estimated values of total Plant Available Nitrogen (PAN) from the sludge applied and that present in the soil for years from 1984 through 1993. Estimated excess nitrogen present in the soil due to sludge application is presented in Table 5. This is the best estimate that could be made without extensive soil sampling. It is expected that in the years when excess nitrogen was applied some soil accumulations would occur. The estimated excess N in the soil was as high as 826 kg/ha. It can be seen that there were some years (1985-1988) when nitrogen loading rates were in excess of the plant requirements. In 1989, 1992, and 1993, a nitrogen deficit was observed.

Table 2. Nitrogen Data of the Applied Sludge

Years	TS % ^a	TKN mg/kg	NH ₃ -N mg/kg	Org.-N mg/kg	NO ₃ -N mg/kg	PAN ^b mg/kg
1984	3.9	57,000	29,070	27,930	74	34,730
1985	3.21	72,000	34,000	38,000	67	41,667
1986	2.35	60,900	45,150	15,750	440	49,136
1987	2.4	89,166	53,298	35,868	62	60,533
1988	3.4	83,676	28,262	55,414	58	39,403
1989	3.5	52,519	23,118	29,401	60	29,058
1990	3.9	55,865	23,372	32,493	81	29,951
1991	3.3	62,122	27,443	34,679	95	34,474
1992	4.7	51,872	20,395	31,477	31	26,721
1993	5.1	45,226	19,757	25,470	28	24,879

^aFrom Table 1

^bPAN = NH₃-N + 0.2 * Org.-N + NO₃-N

Table 3. Nitrogen Nutrient Loading Rates of the Digested Sludge

Years	Application Rates mt/ha ^a	TKN kg/ha	NH ₃ -N kg/ha	Org.-N kg/ha	NO ₃ -N kg/ha	PAN Sludge ^b kg/ha
1984	11.12	634	323	310	0.83	386
1985	27.57	1985	937	1048	1.8	1148
1986	12.06	734	544	190	5.3	587
1987	15.69	1339	836	563	0.97	950
1988	19.82	1658	560	1098	1.15	781
1989	7.42	389	171	218	0.45	215
1990	15.69	876	367	509	0.91	470
1991	10.76	668	295	373	1.02	371
1992	10.31	535	210	325	0.32	275
1993	4.71	213	93	120	0.13	117

^aFrom Table 1^bNitrogen loading rate for i component = (Sludge content of i component) (Application rate)

Table 4. Estimation of Aggregate Plant Available Nitrogen (PAN)

Years	Sludge Application Rates mt/ha ^a	PAN Sludge kg/ha ^b	PAN Soil kg/ha ^c	Total PAN kg/ha
1984	11.12	386	36	422
1985	27.57	1148	36	1184
1986	12.06	587	36	623
1987	15.69	950	36	986
1988	19.82	781	36	817
1989	7.42	215	36	251
1990	15.69	470	36	506
1991	10.76	371	36	407
1992	10.31	275	36	311
1993	4.71	117	36	153

^aFrom Table 1^bFrom Table 2^c[29]

Table 5. Estimation of Excess Nitrogen in the Soil

Years	Sludge Application Rates mt/ha ^a	Total PAN kg/ha ^b	N Uptake by Corn and Wheat kg/ha/yr ^c	Excess N kg/ha
1984	11.12	422	358	9
1985	27.57	1186	358	826
1986	12.06	623	358	265
1987	15.69	986	358	628
1988	19.82	817	358	459
1989	7.42	251	358	None
1990	15.69	506	358	148
1991	10.76	407	358	49
1992	10.31	311	358	None
1993	4.71	153	358	None

^aFrom Table 1

^bFrom Table 3

^cFor corn yield 10.1-11.8 mt/ha, N uptake = 258 kg/ha.

For wheat yield > 46 mt/ha, N uptake = 100 kg/ha.

For soybeans yield > 4.6 mt/ha, N uptake = 336 kg/ha.

Total N uptake by corn and wheat = 258 + 100 = 358 kg/ha.

Total N uptake by corn and soybeans = 258 + 336 = 594 kg/ha.

Water Quality of Groundwater

There was practically no excess nitrogen supplied to the fields due to the sludge application in 1984 (Table 5). Therefore, the nitrate concentrations in the groundwater measured in 1984 were assumed to be background levels. The average nitrate nitrogen level in the groundwater for that year was 0.77 mg/L.

The nitrate nitrogen concentration in all wells over the period of sludge injection operation, Figure 2, were close to the background levels. However, there were a few occasions when increased concentrations of the nitrates occurred. The maximum nitrate nitrogen concentration of 15.8 mg/L that occurred in the groundwater in well No. 1 at the Plant sites was much higher than background levels indicating leaching may have occurred. The nitrate nitrogen levels in well No. 1 exceeded the 10 mg/L limit set by the SDWA twice in 1991 and twice in 1993, and exceeded only once in well No. 2 in 1993. There was no exceedance of nitrate nitrogen limits in wells No. 3 and 4 (Figure 2). It is also not clear why wells No. 1 and 2 had nitrate nitrogen exceedances while wells No. 3 and 4 did not. There was no correlation of groundwater nitrate levels with the excess N applied to the fields (see Table 5). In 1993, there was no excess N applied to the

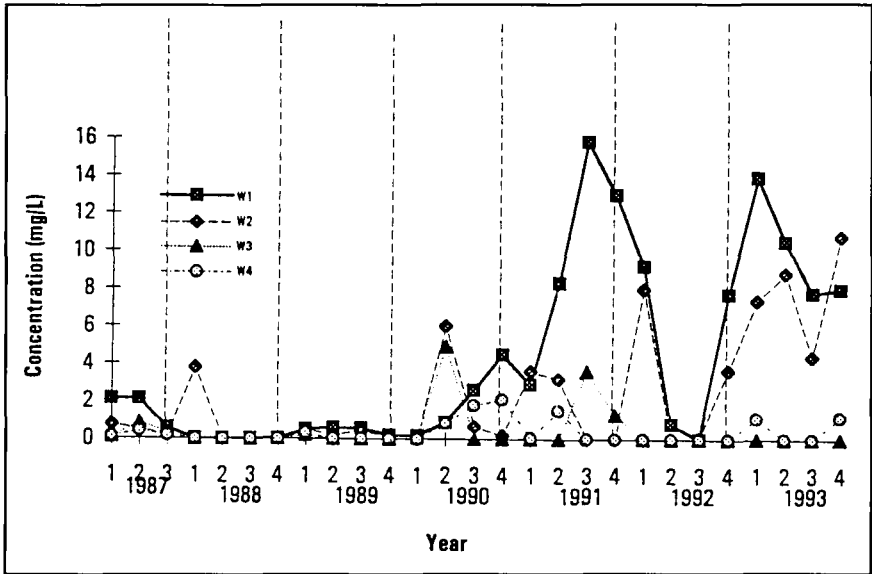


Figure 2. Nitrate-nitrogen levels in groundwater.

soil, nevertheless a nitrate exceedance of the groundwater was observed. There also was no direct correlation of nitrate nitrogen exceedances with the seasons as well. In 1991, these exceedances occurred in September and November, relatively low rainfall months, while in 1993 these occurred in February and May, when generally the groundwater table is higher.

Excess nitrogen applied to the fields essentially has a potential of leaching as $\text{NO}_3\text{-N}$ passes through the soil matrix toward the groundwater table, if the appropriate conditions such as soil permeability, moisture content of the soil, degree of soil aeration, etc., prevail. In low permeable soils such as Menfro silt loam and Carlow silty soil, the migration of nitrate into the groundwater is quite slow [30]. Thus, the nitrates generated by the excess nitrogen supplied to the soil may have leached slowly over a longer period of time and reached the groundwater in 1991 and 1993. The depth to the groundwater table varied from 12 to 18 ft. depending on the season. In flooded conditions the groundwater table was only 2 to 3 ft. from the surface.

In general, there was no evidence of increase in nitrate levels in the groundwater except on the few occasions in 1991 and 1993 due to excess N applied to the fields. This suggests that for the most part, the excess N might have been denitrified and escaped as N_2 from the soil matrix to the atmosphere. This contention has also been supported by the research done by Blanchar et al. [30] at the Plant sites in 1984/1985.

According to Blanchar et al., nitrogen added to the Menfro silt loam soil was found near the surface with a maximum nitrate N content at a depth of about 0.6 m [30]. At the Plant sites in 1984-1985, it was found that a sludge application rate of 18 mt/ha/yr added 1024 kg/ha/yr of total N with excess N of 570 kg/ha/yr to the soil. Of the total N added, 35 percent was removed by the corn and wheat crops, 23 percent remained in the soil, and 43 percent was lost presumably by nitrification and denitrification processes. During the first year of the observations, no N as nitrate or ammonia was found within the profile. They also found that an increased sludge application rate of 72 mt/ha/yr added 4100 kg/ha/yr of total N with an excess of 2282 kg/ha/yr for the same period. Of the total N applied, 13 percent was removed by the crops, 27 percent remained in the soil, and 60 percent was lost through nitrification and denitrification. Even with this excessive application of N, none leached out below 1 m and the increase of nitrate and ammonia in the soil was very small. No large accumulations of ammonia or nitrate were found in the Menfro silt loam soil treated with the sludge application rates from 18 to 72 mt/ha/yr.

Therefore, it can be concluded that the long retention time of nitrates in Menfro silt loam soil and Carlow silt clay soil profiles due to slow water movement along with poor aeration would insure conditions where excess nitrates in the soil at the sites would have been denitrified and not accumulated within the soil profile or leached out of it.

Heavy Metal Loadings

Table 6 shows the annual average concentrations of heavy metals in the applied sludge. The average concentration for each of these metals was below the pollutant concentration limits established in 40 CFR part 503 (Table 7). It can be seen from Table 6 that there were only minor variations in metal concentrations in the digested sludge for the past ten years. The Cu and Zn concentrations were higher than other metals. Copper concentrations in the sludge varied from 610 mg/kg to 1206 mg/kg. Zinc concentrations varied from 914 mg/kg to 1500 mg/kg. Arsenic and cadmium concentrations in the sludge were very low, averaging 5.25 and 6.16 mg/kg, respectively. Table 8 shows the heavy metals loading rates applied to the fields in terms of kg/ha. It can be seen that these cumulative metal loading rates are far less than the EPA limits (Table 7) and there has been no concern about metal buildup in the soils of the application fields.

Most of the metal concentrations in the groundwater monitoring wells were lower than the detectable limits except for Zn, which had high values (varied from 150 to 778 $\mu\text{g/L}$) in June 1989 for all four wells, but these values were still less than that allowable under USEPA Safe Drinking Water Act (SDWA) Secondary Standards.

Table 6. Heavy Metals Data of the Applied Sludge

	TS ^a %	PO ₄ as P mg/kg	As mg/kg	Cd mg/kg	Cr mg/kg	Cu mg/kg	Pb mg/kg	Ni mg/kg	Zn mg/kg
1984	3.9	20,000	6.52	8.8	85	1206	162	96	1,299
1985	3.21	20,000	3.86	4.9	59	775	144	59	914
1986	2.35	22,933	2.6	8.1	46	1050	310	81	1,500
1987	2.4	20,390	< 10	5.5	78	972	209	73	1,251
1988	3.4	—	< 10	4.2	36	732	135	38	938
1989	3.5	10,822	< 3.1	6.3	75	825	214	62	1,278
1990	3.9	21,300	< 0.5	6.7	79	799	184	66	1,339
1991	3.3	9,667	10.8	5.8	63	610	161	79	1,163
1992	4.7	18,475	4.4	6.1	70	785	197	56	1,459
1993	5.1	19,033	< 5.0	5.2	63	709	147	50	1,252

*From Table 1

Table 7. Numerical Criteria for Heavy Metals for 40 CFR Part 503 Rules for Sewage Sludge Land Application

Pollutant	Limits			
	Pollutant Concentration mg/kg	Ceiling Concentration mg/kg	Cumulative Pollutant Loading Rates (CPLR)	
			kg/ha	lb/ac
Arsenic	41	75	41	37
Cadmium	39	85	39	36
Chromium	1,200	3,000	3,000	2,800
Copper	1,500	4,300	1,500	1,300
Lead	300	840	300	270
Mercury	17	57	17	15
Molybdenum	18	75	18	16
Nickel	420	420	420	370
Selenium	36	100	100	89
Zinc	2,800	7,500	2,800	2,500

Table 8. Heavy Metals Loading Rates

	Application Rate ^a dry mt/ha	Loading Rate					
		Cd kg/ha	Cr kg/ha	Cu kg/ha	Pb kg/ha	Ni kg/ha	Zn kg/ha
1984	11.12	0.1	0.95	13.41	1.8	1.1	14.44
1985	27.57	0.14	1.63	21.36	3.97	1.62	25.2
1986	12.06	0.1	0.55	12.66	3.74	0.98	18.09
1987	15.69	0.090	1.22	15.26	3.28	1.14	19.63
1988	19.82	0.080	0.72	14.50	2.68	0.75	18.58
1989	7.42	0.050	0.56	6.12	1.59	0.46	9.48
1990	15.69	0.110	1.24	12.54	2.89	1.03	21.02
1991	10.76	0.060	0.67	6.57	1.74	0.85	12.51
1992	10.31	0.060	0.72	8.09	2.03	0.58	15.04
1993	4.71	0.020	0.29	3.34	0.69	0.24	5.90

Soil Test Data

Tables 9 and 10 show the test results for the soil in the sludge injection site for 1992 and 1993. The metal concentrations for 1993 were slightly higher for some metals in comparison with those for 1992. However, the total P and TKN values were lower in 1993 compared to 1992. There was no indication of heavy metals buildup in the soils from sludge application.

Soil, Sludge, and Groundwater Quality Studies

The wastewater sludge quality as measured in the MU laboratory compared well with the data obtained by the Plant (Tables 2 and 8). The bacteriological quality of the sludge (fecal coliform density of 48,600/g of solids) placed it in EPA Class B category as far as pathogen reduction requirements were concerned.

The data collected by MU personnel for soil, sludge, and groundwater quality at the sites are shown in Table 11. The S-soil samples were collected from 0 to 5 cm from the surface, D-soil samples were collected at deeper sections (20 to 25 cm from top layer), and BG samples were background surface samples collected in nearby field where no sludge has been injected. It can be seen that in most cases the deeper soil layer had lower metal concentrations, except for Cu level in June 1993 S3 sample. This data agrees well with the findings of Dowdy et al. [33]. The metal concentrations in the soil were only somewhat higher than the background soil sample concentrations, except for Cu and Pb levels in June 1993 samples. The soil metal concentrations as determined by MU laboratory

Table 9. 1992 Soil Test Data Summary

Field No.	pH	Conductivity mmhos/cm	TKN mg/kg	P kg/ha	As mg/kg	Cd mg/kg	Cu mg/kg	Pb mg/kg	Ni mg/kg	Zn mg/kg
SC-1	6.3	0.4	1,410	269	9.8	0.21	3.37	17	16	5.0
SC-2	6.2	0.4	1,320	241	4.6	0.34	2.79	15	23	3.8
SC-3	6.1	0.4	1,520	379	4.7	<0.20	7.34	17	17	8.0
SC-4	5.5	0.4	965	276	4.0	0.28	3.65	18	13	5.0
SC-5	6.1	0.4	1,200	175	4.4	<0.20	2.07	16	15	2.9
SC-6E	5.8	0.4	1,580	244	5.3	0.23	3.67	18	15	4.5
SC-6W	6.1	0.5	2,580	344	<4.0	0.36	6.84	23	21	9.2
SC-7E	5.7	0.2	1,760	316	14.0	0.47	5.54	22	14	6.4
SC-7W	5.7	0.3	1,540	321	16.0	0.41	5.84	22	14	6.6
SC-8E	5.9	0.5	2,170	363	14.0	0.48	7.59	22	16	11.4
SC-8W	5.3	1.4	1,520	313	14.0	0.50	8.58	22	14	12.4
SC-9	5.6	0.3	1,150	274	15.0	0.44	5.10	18	18	6.8
SC-10	6.8	0.2	1,220	247	12.0	0.43	3.78	16	15	5.8
SC-11	7.2	0.5	895	126	<4.0	<0.20	1.46	11	19	1.6
SC-12	7.3	0.5	564	122	<4.0	<0.20	1.11	14	19	1.8
Average	6.1	0.5	1,426	267	—	0.38	4.58	18.1	16.6	6.1

Table 10. 1993 Soil Test Data Summary

Field No.	pH	Conductivity mmhos/cm	TKN mg/kg	P kg/ha	As mg/kg	Cd mg/kg	Cu mg/kg	Pb mg/kg	Ni mg/kg	Zn mg/kg
SC-1	6.1	0.3	1,260	234	< 4.0	0.31	17.98	18	14	6.9
SC-2	6.1	0.3	971	224	< 4.0	0.49	6.58	20	18	7.4
SC-3	6.3	0.3	1,140	192	< 4.0	0.32	6.94	22	14	6.2
SC-4	5.6	0.4	1,650	278	< 4.0	0.43	17.09	25	18	12.5
SC-5	6.2	0.4	1,070	193	< 4.0	0.20	6.60	20	15	8.5
SC-6E	6	0.3	1,920	237	< 4.0	0.32	11.21	25	16	10.7
SC-6W	6.2	0.3	1,900	226	< 4.0	0.20	15.35	17	11	11.7
SC-7E	5.4	0.3	1,340	254	< 4.0	0.25	9.62	24	15	8.6
SC-7W	5.4	0.3	690	284	< 4.0	0.50	14.38	26	16	11.8
SC-8E	5.9	0.3	1,210	244	< 4.0	0.41	9.00	25	15	11.6
SC-8W	5.9	0.3	2,000	270	< 4.0	0.47	16.59	28	15	12.4
SC-9	5.6	0.3	1,110	229	< 4.0	0.30	6.40	23	15	8.8
SC-10	6.5	0.4	1,050	206	< 4.0	0.25	6.73	19	14	7.0
SC-11	7.2	0.5	895	126	< 4.0	< 0.20	1.46	11	19	1.6
SC-12	7.3	0.5	564	122	< 4.0	< 0.20	1.11	14	19	1.8
Average	6.1	0.3	1,251	221	< 4.0	0.34	9.80	21.1	15.6	8.5

Table 11. Soil, Sludge, and Groundwater Data for Land Application Site (MU Laboratory Test Data)

SOIL	pH	Conductivity	TS	F.C. ^a		F.S. ^a		NO ₃	Cd	Cr	Cu	Ni	Pb	Zn	As
				%	colonies/g	colonies/g	mg/kg								
Jun-93															
S1	6.13	138	75.5	360	15	20.3	0.55	30.5	6.0	62.0	9.80	24.0	3.27		
S2	6.01	109	76.4	820	37	14.7	0.15	37.5	7.6	77.5	6.40	27.0	3.01		
S3	5.97	106	75.3	540	69	27.4	1.05	24.0	56.0	72.5	29.40	29.5	1.37		
D1	6.97	72	80.2	980	11	9.3	0.50	25.0	6.8	21.0	4.70	19.0	2.16		
BG	5.15	113	78.6	920	23	12.5	ND	13.5	7.0	41.5	9.10	16.5	2.02		
Feb-94															
S1	6.12	89	80.3	254	162	16.1	0.15	26.0	8.0	15.5	28.5	—	2.8		
S2	6.23	60	75.8	130	102	10.2	0.50	21.5	4.5	11.0	19.5	—	1.7		
S3	5.72	64	81.9	51	72	13.1	1.15	20.0	5.0	14.5	29.0	—	1.0		
S4	6.1	63	76.1	152	134	10.7	0.90	13.0	7.0	17.5	24.0	—	1.5		
D1	6.22	103	73.6	300	36	16.1	ND	11.8	8.0	14.0	26.0	—	2.6		
D2	6.45	74	78.0	72	58	7.7	ND	16.5	3.0	10.5	9.5	—	1.6		
BG	7.18	112	71.0	48	40	10.9	ND	14.0	2.0	7.0	15.0	—	0.9		
SLUDGE	7.39	7230	1.32	48,600	41,300	84.5	2.00	15.2	310	121.0	53.5	—	1.9		
GW ^b															
	SU		%	MPN/100 ml											
#1B	6.74	330	—	< 20	< 20	5.2	ND	ND	ND	ND	ND	ND	ND	—	6.6
#4B	6.06	360	—	< 20	< 20	1.9	ND	ND	ND	ND	ND	ND	ND	—	3.2
WW ^b	7.32	1480	—	380,000	330,000	0.5	0.01	0.1	0.3	0.1	0.1	0.1	0.1	—	24.8

^aF.C. = Fecal Coliform; F.S. = Fecal Streptococci, ND = Not Detected

^bGW = Groundwater, WW = Raw Wastewater

were the same order of magnitude as those reported by the Plant (Tables 9 and 10). The nitrate concentrations in the soil were moderate, being only slightly higher than background levels. Bacteriological soil data also indicated that the levels of fecal coliform and fecal streptococci were close to background levels with no evidence of sludge contributing to fecal coliform increases.

The groundwater quality of the monitoring wells showed nitrate nitrogen levels in the range of 1.9-5.2 mg/L which was lower than the SDWA allowable limit (10 mg/L). The metal concentrations in the groundwater samples were below detectable limits except for Arsenic (As) which was well below SDWA permissible limit of 30 µg/L. Thus, there were no detectable metal or nitrate contamination of the groundwater near the sludge application field up to the time of sampling.

SUMMARY AND CONCLUSION

The results of the present study indicate that metal concentrations in groundwater remained low, and in many cases, lower than the detectable limits for the two sludge application sites. For most cases, the bacteriological levels in sludge, soil, and groundwater were well below permissible limits with no evidence of contamination. The results also indicate that there is no heavy metals buildup in sludge-amended soils. However, in some instances the groundwater nitrate nitrogen concentrations were slightly above the 10 mg/L level due to excessive nutrient loadings. In order to reduce the potential risk of further nitrate nitrogen leaching and contamination of the groundwater, the sewage sludge application should follow the agronomic rate such that no excess nitrogen is available for leaching. Land application of sewage sludge is a viable option of recycling/disposing sludge but planning and management is needed to apply sludge at agronomic rates to prevent potential groundwater contamination and soil buildup.

REFERENCES

1. US EPA, *Federal Register*, 40 CFR Part 503, Standards for the Use or Disposal of Sewage Sludge: Final Rules, Washington, D.C., 1993.
2. J. R. Stukenberg, L. W. Jacobs, S. Carr, and S. Bohm, *Long-Term Experience of Sludge Land Application Programs*, Project 91-ISP-4, Water Environment Research Foundation, 1993.
3. US EPA, *Environmental Regulations and Technology Control of Pathogens and Vector Attraction in Sewage Sludge*, EPA/625/R-92/013, Office of Research and Development, Washington, D.C., 1992.
4. T. D. Hinesly, R. L. Jones, J. J. Tyler, and E. L. Ziegler, Soybean Yield Responses and Assimilation of Zn and Cd from Sewage Sludge-Amended Soil, *Journal Water Pollution Control Federation*, 48, pp. 2137-2152, 1976.

5. M. C. Lutrick, The Nutrient Status of Six Field Crops Grown on Soils Treated with Liquid-Digested Sludge, *Soil Crop Science Society, Florida Proceedings*, 33, pp. 183-185, 1974.
6. C. Lamb, Sewage Sludge Produces Hay, *Environmental Action Bulletin*, 3:21, pp. 4-5, 1972.
7. L. W. Jacobs, Agricultural Application of Sewage Sludge, in *Sludge and Its Ultimate Disposal*, J. A. Borchardt, W. J. Redman, G. E. Jones, and R. T. Sprague (eds.), Ann Arbor Science Publishers, Inc., Ann Arbor, Michigan, pp. 109-126, 1981.
8. C. J. Santhanam and D. Schiff, Ultimate Disposal of Sludges, in *Sludge Treatment*, W. W. Eckenfelder, Jr. and C. J. Santhanam (eds.), Marcel Dekker, Inc., New York, pp. 475-522, 1981.
9. D. R. Keeny, Sources of Nitrate to Groundwater, in *Nitrogen Management and Groundwater Protection*, R. F. Follett (ed.), Elsevier Science Publishers, Amsterdam, The Netherlands, pp. 23-34, 1989.
10. Council for Agricultural Science and Technology (CAST), *Agriculture and Groundwater Quality*, Council for Agricultural Science and Technology Report 103, Ames, Iowa, 62P, 1985.
11. G. A. Peterson and W. W. Frye, Fertilizer Nitrogen Management, in *Nitrogen Management and Groundwater Protection*, R. F. Follett (ed.), Elsevier Science Publishers, Amsterdam, The Netherlands, pp. 183-220, 1989.
12. T. D. Hinesly, O. C. Braids, and J. A. E. Molina, *Agricultural Benefits and Environmental Changes Resulting from the Use of Digested Sewage Sludge on Field Crops*, A Report on a Solid Waste Demonstration Project, US EPA, Washington, D.C., 1971.
13. T. K. Soon, T. E. Bates, E. G. Beauchamp, and J. R. Mayer, Land Application of Chemically Treated Sewage Sludge: I. Effects on Crop Yield and Nitrogen Availability, *Journal of Environmental Quality*, 7, pp. 264-269, 1978.
14. N. E. Stewart, E. G. Beauchamp, C. T. Croke, and L. R. Webber, Nitrate Nitrogen Distribution in Corn Land following Applications of Digested Sewage Sludge, *Journal of Soil Science*, 55, pp. 287-294, 1975.
15. T. M. Addiscott, A. P. Whitmore, and D. S. Powlson, *Farming, Fertilizers and Nitrate Problems*, CAB International, Oxford, England, pp. 31-43, 1991.
16. J. H. A. M. Steenvoorden, Agricultural Practices to Reduce Nitrogen Losses Via Leaching and Surface Runoff, in *Management Systems to Reduce Impact of Nitrates*, J. C. Germon (ed.), Elsevier Science Publishers LTD, Essex, England, 1989.
17. B. E. Reed, P. E. Carrier, and M. R. Matsumoto, Applying Sludge on Agricultural Land, *Biocycle*, pp. 58-61, July 1991.
18. CAST, *Effects of Sewage Sludge on the Cadmium and Zinc Content of Crops*, Council for Agricultural Science and Technology, Ames, Iowa, 1980.
19. R. L. Chaney, J. R. Heckman, and J. S. Angle, Residual Effects of Sewage Sludge on Soybean: I. Accumulation of Heavy Metals, *Journal of Environmental Quality*, 16, pp. 113-118, 1987.
20. F. C. Boswell, Municipal Sewage Sludge and Selected Element Application to Soil: Effect on Soil and Fescue, *Journal of Environmental Quality*, 4, pp. 267-273, 1975.
21. L. Yangming and R. B. Corey, Redistribution of Sludge-Borne Cadmium, Copper, and Zinc in a Cultivated Plot, *Journal of Environmental Quality*, 22, pp. 1-8, 1993.

22. A. C. Chang, A. L. Page, K. W. Foster, and T. E. Jones, A Comparison of Cadmium and Zinc Accumulation by Four Cultivars of Barley Grown in Sludge-Amended Soils, *Journal of Environmental Quality*, 11, pp. 409-412, 1982.
23. R. H. Dowdy and V. V. Volk, Movement of Heavy Metals in Soil, in *Chemical Mobility and Reactivity in Soil Systems*, D. W. Nelson et al. (eds.), SSSA, Madison, Wisconsin, pp. 229-240, 1983.
24. P. R. Fresquez, R. E. Francis, and G. L. Dennis, Sewage Sludge Effects on Soil and Plant Quality in a Degraded, Semiarid Grassland, *Journal of Environmental Quality*, 19, pp. 324-329, 1990.
25. W. K. Robertson, M. C. Lutrick, and T. L. Yuan, Heavy Application of Liquid-Digested Sludge on Three Ultisols: I. Effects on Soil Chemistry, *Journal of Environmental Quality*, 11, pp. 278-282, 1982.
26. D. C. Martens, B. D. Rappaport, R. B. Reneau, Jr., and T. W. Simpson, Metal Availability in Sludge Amended Soils with Elevated Metal Levels, *Journal of Environmental Quality*, 17, pp. 42-47, 1988.
27. A. C. Chang, W. E. Emmerich, L. J. Lund, and A. L. Page, Movement of Heavy Metals in Sewage Sludge-Treated Soils, *Journal of Environmental Quality*, 11, pp. 174-178, 1982.
28. US EPA, *POTW Sludge Sampling and Analysis Guidance Document*, Washington, D.C., 1989.
29. APHA, *Standard Methods for the Examination of Waste and Wastewater* (16th Edition), Washington, D.C., 1985.
30. R. W. Blanchar, P. Koeling, C. L. Scriver, and W. P. Teaque, *Distribution of Sludge Components from the Hinkson-Perche Wastewater Treatment Facility in Soil, Plant, Climate System*, Report to City of Columbia, Missouri, 1985.
31. US EPA, *Process Design Manual for Land Application of Municipal Sludge*, 1983.
32. Missouri Department of Natural Resources (MDNR), *Standard Conditions for NPDES Permits, Part IV, Land Application of Biosolids*, 1993.
33. R. H. Dowdy, J. J. Latterell, T. D. Hinesly, R. B. Grossman, and D. L. Sullivan, Trace Metal Movement in an Aeric Ochraquaf Following 14 Years of Annual Sludge Applications, *Journal of Environmental Quality*, 20, pp. 119-123, 1991.

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