

Soil Carbon Sequestration Resulting from Long-Term Application of Biosolids for Land Reclamation

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Investigations on the impact of application of biosolids for land reclamation on C sequestration in soil were conducted at Fulton County, Illinois, where 41 fields (3.6–66 ha) received biosolids at a cumulative loading rate from 455 to 1654 dry Mg ha⁻¹ for 8 to 23 yr in rotation from 1972 to 2004. The fields were cropped with corn, wheat, and sorghum and also with soybean and grass or fallowed. Soil organic carbon (SOC) increased rapidly with the application of biosolids, whereas it fluctuated slightly in fertilizer controls. The peak SOC in the 0- to 15-cm depth of biosolids-amended fields ranged from 4 to 7% and was greater at higher rates of biosolids. In fields where biosolids application ceased for 22 yr, SOC was still much higher than the initial levels. Over the 34-yr reclamation, the mean net soil C sequestration was 1.73 (0.54–3.05) Mg C ha⁻¹ yr⁻¹ in biosolids-amended fields as compared with -0.07 to 0.17 Mg C ha⁻¹ yr⁻¹ in fertilizer controls, demonstrating a high potential of soil C sequestration by the land application of biosolids. Soil C sequestration was significantly correlated with the biosolids application rate, and the equation can be expressed as $y = 0.064x - 0.11$, in which y is the annual net soil C sequestration (Mg C ha⁻¹ yr⁻¹), and x is annual biosolids application in dry weight (Mg ha⁻¹ yr⁻¹). Our results indicate that biosolids applications can turn Midwest Corn Belt soils from current C-neutral to C-sink. A method for calculating SOC stock under conditions in which surface soil layer depth and mass changes is also described.

STRIP-MINING causes loss of topsoil and destruction of soil structure, leading to low soil organic matter (SOM) and compaction (Peterson et al., 1979). Water quality problems also exist in watersheds of mined land (Soper, 1992). Reclamation of strip-mined land has the potential to increase the agricultural productivity of Illinois soils. Management is often needed for reclamation of strip-mined land because it might take centuries to develop a productive soil from mine spoils left to nature (Smith et al., 1971). To contribute to improving the productivity of the 16,400 ha of mine spoils in Fulton County, Illinois, representing 7.24% of total county land area (Haynes and Klimstra, 1975), the Metropolitan Water Reclamation District of Greater Chicago (MWRDGC) initiated the world's largest land reclamation project with the biosolids in 1972.

The safe and economic disposal of biosolids produced from the treatment of wastewater has been a challenge for municipal wastewater treatment agencies. Although incineration and land-filling of biosolids were used by many municipalities in the early 1970s, the MWRDGC believed that the utilization of the fertilizer value of municipal biosolids offers the best alternative to municipal wastewater treatment agencies. The process of utilization rather than disposal requires relatively small amounts of energy and offers to farmers a source of free or inexpensive fertilizer. Data from monitoring reported by Tian et al. (2006) indicated that the long-term application of high rates of biosolids produced before and after the promulgation of federal regulation had only a minor impact on surface water quality. Granato et al. (2004) noted that the transfer of trace metals from biosolids to corn grain was less than that predicted in the risk model of USEPA's 40 CFR Part 503 regulation, which was promulgated in 1993 to govern the land application of biosolids in the USA. With the increased economic restraints and environmental concerns about land-filling and incineration, interest in land application could continue to grow (O'Connor et al., 2005). Because of the high organic matter content, biosolids are ideal for reclaiming degraded soils by increasing organic matter and improving the structure.

The global SOC inventory is estimated to be 1200 to 1600 Pg, which is close to the combined amounts stored in terrestrial vegetation (550–700 Pg) and the atmosphere (750 Pg) (Post et al., 1990;

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Abbreviations: MWRDGC, Metropolitan Water Reclamation District of Greater Chicago; SOC, soil organic carbon; SOM, soil organic matter; WAS, waste-activated sludge; WRPs, water reclamation plants.

Sundquist, 1993). Therefore, even a small percentage change in the SOC pool could easily affect the change in atmospheric CO₂. Increasing soil C reserves in agricultural and rangeland soils and restoring degraded soils to productivity have been considered as an important means to sequester C (Lal et al., 1998). Specifically, Jarecki and Lal (2003) recommended the use of organic waste materials as an important management practice that would help fill the large C sink in the world's agricultural soils. We hypothesize that the application of biosolids results in C sequestration in soil through increasing soil microbial biomass, an important SOM source, and that the amorphous Fe and Al oxides present in biosolids promote the humification of organic residues, also contributing to soil C sequestration.

Although there are many reports on the long-term change in SOM, most of the previous studies reported measurements usually at the beginning and at the end of the study period, lacking repeated measurements of results throughout the studies. Long-term estimates of soil C sequestration still rely almost exclusively on modeling (Jones and Donnelly, 2004). To fill some of the gaps in long-term dynamics of SOM with the addition of organic amendments and to quantify the soil C sequestration by biosolids land application, this report presents SOC concentration dynamics and C sequestration in soil that received biosolids for over 30 yr for reclaiming strip-mined land.

Materials and Methods

Study Site

The study site is located at Fulton County in western Illinois, approximately 300 km southwest of Chicago. The site has a continental climate, with an annual mean air temperature of 10.4°C and annual precipitation of 1013 mm. The monthly mean surface soil temperature and moisture ranged from 1 to 25°C and 13 to 27%, respectively (WARM, 2008). The Fulton county soil environment is influenced by an extended wet and cold winter; a short, wet, and cool spring; a dry, hot summer; and a moist, cool fall. As Gilmour and Gilmour (1980) suggested 20% as optimal soil moisture and 25°C as optimal soil temperature for biosolids decomposition, low temperature for most months of the year at Fulton County leads to slower biosolids decomposition, though the waterlogging in the colder months and moisture deficiency in warmer months are also unfavorable to microbial activity. Land in the study site was strip-mined in the early 1900s, resulting in some of the previously productive agriculture areas becoming roughly scarred wastelands. The pH of surface spoils was neutral to alkaline, and the texture varies mostly from silt loam to silty clay loam.

Treatment Establishment

In the 6000 ha of land comprising calcareous strip-mined and nonmined soil, acidic coal refuse materials, mine lakes, and wooded areas, approximately 1790 ha of calcareous strip-mined and nonmined low-productivity soil was developed into 80 fields, 58 of which were used for biosolids application. Among 58 fields for the project, 16 fields received only supernatant from liquid biosolids holding basins, and the amount of solids added

(0.37–21.5 dry Mg ha⁻¹ for the period of 33 yr) through such applications was low enough to be negligible. One field was also excluded from this study because it received biosolids for only 4 yr. This study therefore included the 41 fields as listed in Table 1. The fields were sized from 3.6 to 66 ha and received biosolids at cumulative loading rates of 455 to 1654 dry Mg ha⁻¹ for 8 to 23 yr in rotation from 1972 to 2004.

The biosolids-amended fields were divided into three groups based on their major soil types (Table 1). Group I consists of 20 fields of mine spoil soils, primarily Lenzburg and Lenzwheel soil series: "fine-loamy, mixed, active, calcareous, mesic Alfic Udarents" (USDA-NRCS, 1997). Soils in these fields were derived from overburden from surface mining, consisting of unconsolidated materials, which include solum and substratum of the premined soil, and consolidated materials, which include shale or sandstone bedrock. Group II consisted of nine fields of "fine" mine spoil, primarily Rapatee soil series: "fine-silty, mixed, superactive, nonacid, mesic Alfic Udarents." The Rapatee soils also derived from overburden from surface mining, but in areas of these soils, the topsoil and subsoil were saved during the mining and redistributed on graded rocky overburden after the mining. Therefore, soils in Group II fields had a "finer" surface layer than those in Group I at the time of the reclamation project. Group III consisted of 12 fields of nonmined soil of various series (Table 1). Although soils in this group were not mined, they were degraded by intensive cultivation and/or overgrazing.

The control we selected for this study is the fields that received chemical fertilizer and had a cropping system similar to biosolids-amended fields. The biosolids-amended fields were cropped with rotations of cereal crops such as corn, wheat, and sorghum. Soybean, grass, and fallow were also used in these fields. The supernatant fields and two unamended fields (fields F24 and F38) were generally unfertilized and were used for grass, although some of these fields were cropped with corn or soybean during the most recent decade. Among all unamended fields, only two fields (F18 and F29) regularly received commercial fertilizer and had a cropping system similar to biosolids-amended fields; therefore, F18 and F29 served as controls. Field 29 (Lenzburg soil) received only one biosolids application at 1.21 Mg ha⁻¹ in 1979 and served as control for Group I. The nonmined half of F18 (F18-2) received only one biosolids application at 1.04 Mg ha⁻¹ in 1979 and was used as control for Group III. No field was suitable as a control for Group II, but we used F18-2 as a reference because the "fine" surface layer in Group II was mainly premined soil, as in F18-2.

Application of Biosolids

The biosolids applied to the fields were generated at the MWRDGC's Stickney and Calumet water reclamation plants (WRPs). The sewage at these treatment plants includes domestic and industrial wastewater at a ratio of 3:2 plus rainwater. Large objects and grits in the sewage are removed and sent to a landfill before the sewage proceeds to a sediment tank to obtain primary sludges and to an aeration tank for the waste-activated sludge (WAS). Primary sludges mainly consist of fecal solids, and WAS is the active biological material and is largely composed of sapro-

Table 1. List of fields of three groups used for biosolids reclamation of strip-mined and nonmined degraded soils at Fulton County, Illinois.

Field no.	Location		Soil taxonomy	FLD area	Cum biosolids appl. rate	No of appl. yrs	Yrs since last appl
	Latitude	Longitude					
Group I: mine spoil soil							
F29 (Fert CL)	40°27'44"N	90°06'42"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	12.2	1.2	1	27
F32	40°27'33"N	90°05'02"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	3.6	602	11	2
F39	40°32'28"N	90°05'35"W	Lenzwheel silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	12.4	646	11	2
F25	40°28'49"N	90°03'43"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	15.4	764	13	6
F27	40°28'20"N	90°03'30"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	15.2	847	16	4
F28	40°28'12"N	90°04'26"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	11.1	903	12	4
F26	40°28'22"N	90°03'53"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	25.9	982	16	6
F13	40°29'11"N	90°05'47"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	9.8	1068	17	2
F12	40°28'55"N	90°06'06"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	10.9	1093	17	6
F33	40°27'43"N	90°04'53"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	6.2	1110	12	2
F8	40°31'09"N	90°05'54"W	Lenzwheel silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	18.2	1131	18	4
F30	40°27'51"N	90°04'06"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	27.9	1169	15	6
F11	40°29'20"N	90°06'34"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	8.5	1180	16	6
F14	40°29'23"N	90°05'32"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	20.0	1184	20	2
F4	40°30'42"N	90°05'34"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	38.9	1216	19	4
F15	40°29'45"N	90°04'22"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	15.7	1238	16	2
F5	40°31'03"N	90°05'42"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	8.3	1361	20	3
F7	40°31'22"N	90°06'04"W	Lenzwheel silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	42.2	1385	19	4
F3	40°30'10"N	90°06'00"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	16.6	1420	22	2
F1	40°29'37"N	90°06'33"W	Lenzwheel silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	16.0	1428	19	2
F2	40°29'53"N	90°06'02"W	Lenzburg silt loam: Fine-loamy, mixed, active, calcareous, mesic Alfic Udarents	19.0	1654	20	4
Group II: "fine" mine spoil soil (mine spoil soil + pre-mined soil)							
F40	40°32'03"N	90°05'34"W	(1/2) Rapatee silty clay loam: Fine-silty, mixed, superactive, nonacid, mesic Alfic Udarents and (1/2) Sable silty clay loam: Fine-silty, mixed, superactive, mesic Typic Endoaquolls	27.5	497	8	23
F44	40°32'54"N	90°04'44"W	Rapatee silty clay loam: Fine-silty, mixed, superactive, nonacid, mesic Alfic Udarents	12.0	805	12	2
F45	40°33'28"N	90°05'31"W	Rapatee silty clay loam: Fine-silty, mixed, superactive, nonacid, mesic Alfic Udarents	46.7	845	13	3
F42	40°32'38"N	90°05'16"W	Rapatee silty clay loam: Fine-silty, mixed, superactive, nonacid, mesic Alfic Udarents	65.6	858	13	3
F41	40°32'42"N	90°05'56"W	Rapatee silty clay loam: Fine-silty, mixed, superactive, nonacid, mesic Alfic Udarents	17.5	979	12	2
F43	40°32'55"N	90°05'34"W	Rapatee silty clay loam: Fine-silty, mixed, superactive, nonacid, mesic Alfic Udarents	21.0	983	11	2
F47	40°33'07"N	90°04'58"W	Rapatee silty clay loam: Fine-silty, mixed, superactive, nonacid, mesic Alfic Udarents	9.4	1121	12	2
F9	40°30'55"N	90°06'32"W	Rapatee silty clay loam: Fine-silty, mixed, superactive, nonacid, mesic Alfic Udarents	26.7	1269	22	3
F17	40°30'45"N	90°06'09"W	Rapatee silty clay loam: Fine-silty, mixed, superactive, nonacid, mesic Alfic Udarents	27.7	1417	23	3
Group III: Nonmined soil							
F18-2 (Fert CL)†	40°27'51"N	90°06'22"W	Sable silty clay loam: Fine-silty, mixed, superactive, mesic Typic Endoaquolls	22.7	1.0	1	27
F22	40°28'38"N	90°04'05"W	Clarksdale Silt loam: Fine smectitic, mesic Udollic Endoaqualls	11.4	455	13	22
F23	40°28'28"N	90°05'07"W	Clarksdale Silt loam: Fine smectitic, mesic Udollic Endoaqualls	6.2	473	10	22
F20	40°28'36"N	90°04'09"W	Ipava Silt loam: Fine, smectitic, mesic Aquic Argiudolls	40.9	531	11	22
F31	40°27'25"N	90°04'47"W	Orthents, silty, undulating: Coarse-silty, mixed, superactive, nonacid, mesic Aquic Udifluvents	4.4	557	10	22
F34	40°27'54"N	90°05'04"W	Clarksdale Silt loam: Fine smectitic, mesic Udollic Endoaqualls	22.6	566	10	22
F21	40°28'52"N	90°04'12"W	Greenbush Silt loam: Fine-silty, mixed, superactive, mesic Mollic Hapludalfs	11.3	618	12	22
F19	40°29'11"N	90°03'45"W	Clarksdale Silt loam: Fine smectitic, mesic Udollic Endoaqualls	13.9	644	11	22
F37	40°26'54"N	90°03'05"W	Greenbush Silt loam: Fine-silty, mixed, superactive, mesic Mollic Hapludalfs	24.4	801	12	4
F35	40°27'18"N	90°03'00"W	Rozetta Silt loam: Fine-silty, mixed, superactive, mesic Typic Hapludalfs	48.1	1049	13	4
F10	40°29'12"N	90°06'58"W	Keomah silt loam: Fine, smectitic, mesic, Aerlic Endoaqualls	30.8	1073	14	14
F36	40°27'24"N	90°02'31"W	Ipava Silt loam: Fine, smectitic, mesic Aquic Argiudolls	19.8	1083	12	4
F16	40°29'25"N	90°05'00"W	Rozetta Silt loam: Fine-silty, mixed, superactive, mesic Typic Hapludalfs	60.8	1084	16	6

† Also serve as control for Group II.

‡ Dry weight.

phytic bacteria, protists, and filter-feeding species. The WAS and a small percentage of primary sludges were anaerobically digested at 35°C for at least 15 d to meet the minimum criteria as biosolids. Throughout the project, three types of biosolids (lagooned liquid, lagooned dewatered, and lagooned air-dried biosolids)

were used. From 1971 to 1983, 730,000 Mg (dry weight) of anaerobically digested sludges were barged to Fulton County. Biosolids shipped from 1971 to 1981 had an average of 6% solids and came from digesters and lagoons. From 1981 to 1983, centrifuge-dewatered biosolids containing an average of 25%

solids were delivered to Fulton County (Hall et al., 1985). At the Fulton County land reclamation site, biosolids were stored in three 12-m-deep holding basins before application. With a few months, materials in the holding basins partitioned into a supernatant (total solids = 0.3%) phase and thickened underlying slurry (total solids = 12%). Supernatant was barged back to water treatment plants in Chicago from 1972 to 1976 and applied to “supernatant fields” from 1976. A hydraulic dredge was used to resuspend the slurry in a holding basin and to deliver it to a pump facility for distribution to fields. This slurry contained 4% total solids and was referred to as “liquid biosolids.” Because the liquid biosolids had been stored in holding basins for several months to years before application, more precisely it was the “lagooned liquid biosolids.” As the water level was lowered during the application season, solids at the holding basin bottom were dug out and deposited on the holding basin walls. After air-drying, the solids were removed from the basin walls and applied to fields or stockpiled for additional drying over the winter months. This material contained an average of 47% solids and was referred to as “dewatered biosolids.” Generally, the dewatered biosolids were stored in a holding basin longer than liquid biosolids. Air-dried biosolids were produced in Chicago using a low-cost technique. Anaerobically digested sludges were first stored in a lagoon-like holding basin for a minimum of 18 mo for further stabilization. After that, materials in the lagoon were dug out and air-dried with agitation on a paved bed to contain approximately 60 to 70% total solids. Liquid biosolids were applied from 1972 to 1985 primarily by incorporation discs, although a small portion was applied by a traveling sprinkler during the initial 4 yr. Dewatered biosolids were applied from 1980 to 1995 and air-dried biosolids from 1987 to 2004 by a manure spreader followed by discing incorporation (Skuse et al., 1991). The application of biosolids normally occurred from June to September in fields where crop was harvested in previous years or where wheat was just harvested. Some of the fields receiving biosolids were cropped in the same year; however, many of them were planted with a cover crop (mainly rye). The rye field was sprayed with herbicides and disced in the subsequent year for cropping. The annual biosolids application rates to various fields are shown in Table 2. The MWRDGC’s three decades of land reclamation project at Fulton County applied a cumulative total of 830,000 Mg biosolids to land.

The “40 CFR Part 503” biosolids federal regulation was promulgated in 1993 (USEPA, 1994). The Part 503 rule stipulates concentrations and loading rates of metals for biosolids applied to land. Biosolids with metal concentrations lower than ceiling limits are safe for land application, but application has to be stopped when cumulative applications reach the cumulative loading limits. Biosolids with metal concentrations lower than pollutant limits (i.e., exceptional quality biosolids) can be applied to land indefinitely. Since the promulgation of Part 503, the MWRDGC’s biosolids have met the exceptional quality metal concentration limits. The remaining pre-Part 503 biosolids already shipped to Fulton County were mixed with sand to meet the metal limits for applications only in 1994 and 1995.

Sampling and Analysis

Soil samples were collected at a depth of 0 to 15 cm each spring before the annual biosolids application. In collecting soil samples, each field was divided into two halves. In each half, about 20 to 40 cores, depending on field size, were taken to make one composite sample. The samples from the two halves were further composited into one sample and analyzed in duplicate. All analyses were controlled with check samples to ensure analyses were consistent for a period of three decades. The concentration of SOC was determined by the Walkley-Black method. A composite sample of biosolids applied to each field was collected for analysis. The total solids in biosolids were determined by drying the biosolids at 103 to 105°C. The volatile solids or organic matter in biosolids was estimated as loss in mass at 550°C (USEPA, 1983).

The N in biosolids was determined using Kjeldahl digestion, followed by colorimetric analysis. Total Fe and Al were determined by digestion in nitric acid, followed by analysis using atomic absorption spectrophotometry from 1971 to 1998 and inductively coupled plasma spectroscopy thereafter. A mean of all composite samples in a year was calculated.

Calculation of Soil Carbon Gain

Because biosolids were mixed with only surface soil, we could assume the SOC was unchanged in the subsoil. Therefore, we considered the difference in C stock in the “surface soil layer” between a given year and the first year of the biosolids application as the soil C gain after biosolids land reclamation.

Generally, soil C stock can be calculated using the following formula:

$$Mc = DS\rho C/100 \quad [1]$$

where Mc is the soil C stock (Mg ha^{-1}), D is the surface soil layer depth (m) (0.15 at $t = 0$ in 1972), S is the surface area of a hectare ($10,000 \text{ m}^2 \text{ ha}^{-1}$), ρ is bulk density (Mg m^{-3}), and C is the SOC concentration (%).

The known bulk densities of the 0- to 15-cm surface soil layer at the start of the project ($t = 0$) were 1.61 Mg m^{-3} for the strip-mined soil and 1.29 Mg m^{-3} for the nonmined areas (Peterson et al., 1979). The bulk density of 1.61 Mg m^{-3} was given to most of the mine spoil soil fields and 1.29 Mg m^{-3} to most of the “fine” mine spoil soil and nonmined soil fields. A few fields had extremely high or low initial SOC; therefore, bulk density in these fields was adjusted using the SOC and bulk density equation established from measurements in selected fields in 2005. After the start of biosolids application, the bulk density in biosolids-amended soil was calculated as the weighted mean of soil ρ in the 0.15-m depth and ρ of newly applied biosolids:

$$\rho_t = [(0.15S\rho_{t-1})\rho_{t-1} + W_{BS(t-1)}\rho_{BS}]/(0.15S\rho_{t-1} + W_{BS(t-1)}) \quad [2]$$

where ρ_t is the bulk density of biosolids-amended soil at t , ρ_{t-1} is the bulk density of biosolids-amended soil at $t - 1$, $W_{BS(t-1)}$ is the dry weight of biosolids applied at $t - 1$, and ρ_{BS} is the bulk density of biosolids. The biosolids had a mean bulk density of 0.69 Mg m^{-3} (Granato et al., 2004). We verified such bulk density estimation by measuring soil bulk density in samples

collected from six selected fields in 2005. The calculated mean bulk density was 1.12 (1.02–1.19) Mg m⁻³, as compared with measured bulk density of 1.11 (0.94–1.18) Mg m⁻³, with a median deviation of 0.01 (–0.08 to 0.04) Mg m⁻³ between them. The soil bulk density in the Group I fertilizer control field (F29, Lenzburg soil) was 1.44 g cm⁻³ in 2005, 0.17 Mg m⁻³ less than that in 1972. Therefore, we proportioned such an increment over the entire period. The bulk density in the nonmined portion of F18 that is used as Group II and III control was not included in the 2005 sampling. We considered bulk density in this control unchanged because it had very little change in SOC by 2005.

After each year's biosolids application, there was an increase in the surface soil layer as ΔA_p (Fig. 1), as reported by Chang et al. (2007) for long-term application of animal manure; therefore, there was an additional C stock in the ΔA_p for the biosolids-amended soil, and this can be calculated as follows:

$$Mc_{\Delta A_p} = D_{\Delta A_p} Sp_{\Delta A_p} C_{\Delta A_p} / 100 \quad [3]$$

in which $Mc_{\Delta A_p}$ is the C stock in the ΔA_p at t , $D_{\Delta A_p}$ is the depth of ΔA_p induced by biosolids application at $t - 1$, $\rho_{\Delta A_p}$ is the bulk density of ΔA_p , and $C_{\Delta A_p}$ is the SOC concentration in ΔA_p . We estimated the $D_{\Delta A_p}$ for each year using the following formula:

$$D_{\Delta A_p} = [(0.15Sp_{t-1} + W_{BS(t-1)}) / \rho_t / S] - 0.15 \quad [4]$$

Because the biosolids applied at $t - 1$ that created ΔA_p at t was mixed with the 0- to 15-cm soil by incorporation, the SOC concentration in 0- to 15-cm soil depth (C_{con15}) sampled at t and the bulk density (ρ_{15}) estimated for t can also be considered as that in ΔA_p induced by the biosolids applied at $t - 1$ (i.e., $C_{\Delta A_p} = C_{con15}$; $\rho_{\Delta A_p} = \rho_{15}$).

The ΔA_p accumulated with the continuous application of biosolids, as indicated from $t = 0$ to $t = 34$ in Fig. 1. Therefore, the SOC stock at year t should include those in the 0- to 15-cm depth at t , ΔA_p induced by biosolids application at year $t - 1$, and the sum of all ΔA_p up to year $t - 1$. The decomposition of the SOC in the current ΔA_p has been reflected in the SOC concentration measured in the 0- to 15-cm depth. The loss of SOC in previous ΔA_p depths through decomposition was estimated using the first-order decay equation.

Finally, the SOC stock at t , starting from $t = 0$ with a step of 1 (year), could be estimated by the following formula:

$$Mc = 0.15Sp_{15} C_{con15} / 100 + \sum_{n=1}^t (D_{\Delta A_p} Sp_{\Delta A_p} C_{\Delta A_p} / 100)_n e^{-k(t-n)} \quad [5]$$

where k is the annual decomposition rate of SOC left in ΔA_p , 0.0276 yr⁻¹ or 2.7%, which was obtained from the dynamics of SOC at the 0- to 15-cm depth after the cessation of biosolids applications in this study.

Estimation of Biosolids Carbon Remaining

The decomposition of biosolids follows the first-order double exponential decay (Gilmour and Gilmour, 1980): $y = Re^{-k_1 t} + (1 - R)e^{-k_2 t}$, where R is the rapidly decomposable fraction, k_1 is the decomposition rate constant for R fraction, $(1 - R)$ is the slowly decomposable fraction, and k_2 is the decomposi-

tion rate constant for $(1 - R)$ fraction. Although several biosolids decomposition models are available (Gilmour et al., 1996; 2003), they generally overestimated the biosolids decomposition because these models were built on short-term experiments and did not separate the prime effect (a stimulation of native SOM decomposition due to the addition of organic materials) (Terry et al., 1979a). The optimal soil moisture and temperature in the laboratory also led to overestimation of biosolids decomposition. Based on the estimation of Gilmour and Gilmour (1980), the decomposition of biosolids hypothetically at a site similar to Fulton County's ambient soil moisture and temperature was only 37% of that under optimal soil moisture (20%) and temperature (25°C). Therefore, we used two steps in constructing a model to estimate the biosolids C remaining after the decomposition of biosolids in the field: (i) establishing a decomposition equation using CO₂ evolved from only biosolids under optimal soil moisture and temperature conditions then (ii) adjusting constants of the equation to Fulton County field condition.

Terry et al. (1979b) conducted a unique study on biosolids decomposition because it used ¹⁴C-labeled synthetic biosolids. This study, therefore, could separate the prime effect and obtain the CO₂ evolved from only biosolids during decomposition. Furthermore, the duration of the Terry et al. (1979b) study was relatively long (336 d) compared with other studies. However, this work was under-cited, most probably because their data were fitted to a linear model of CO₂ evolution vs. log of time (Terry et al., 1979a) rather than to the double exponential model. We compared two fittings ("log of time" and "double exponential") using their data and found that the double exponential had lower sum of square. Therefore, we re-fitted their data into a double exponential decay model.

The synthetic sludge made by Terry et al. (1979b) had volatile solids of 52% and organic N of 3.1%, close to the volatile solids (55%) and organic N (3.7%) of our unlagged liquid biosolids. Because Terry et al. (1979b) did not include 25°C (optimal temperature), we calculated the CO₂ evolved from biosolids at 25°C using Q_{10} over the two temperatures of 21 and 30°C tested in Terry et al. (1979b). Then, we adjusted k_1 and k_2 by multiplying 0.37 to obtain $k_1 = 0.0205$ (d⁻¹) and $k_2 = 0.000301$ (d⁻¹). From Terry et al. (1979b), we got $R = 0.339$. Because R is independent of soil environmental condition, it is not necessary to adjust for field condition. However, R has to be adjusted for different types of biosolids.

At MWRDGC, we measured the volatile solids in biosolids subjected to various further stabilizations. The volatile solids were 55.4% for digester drawoff, 52.5% for centrifuge-dewatered cake, 48.4% for a 2-yr lagooned liquid, and 37% for lagooned air-dried biosolids. The volatile solids content in Table 3 therefore indicates that biosolids were already subjected to various degrees of stabilizations before application; hence, it is necessary to take this into consideration in estimating biosolids C remaining. Because the rapid fraction of biosolids decomposes fast once it is mixed with soil, the rapid fraction could be assumed to decompose completely before the decomposition of the slow fraction starts (Gilmour et al., 1996). We can, therefore, consider the variation in volatile solids with applica-

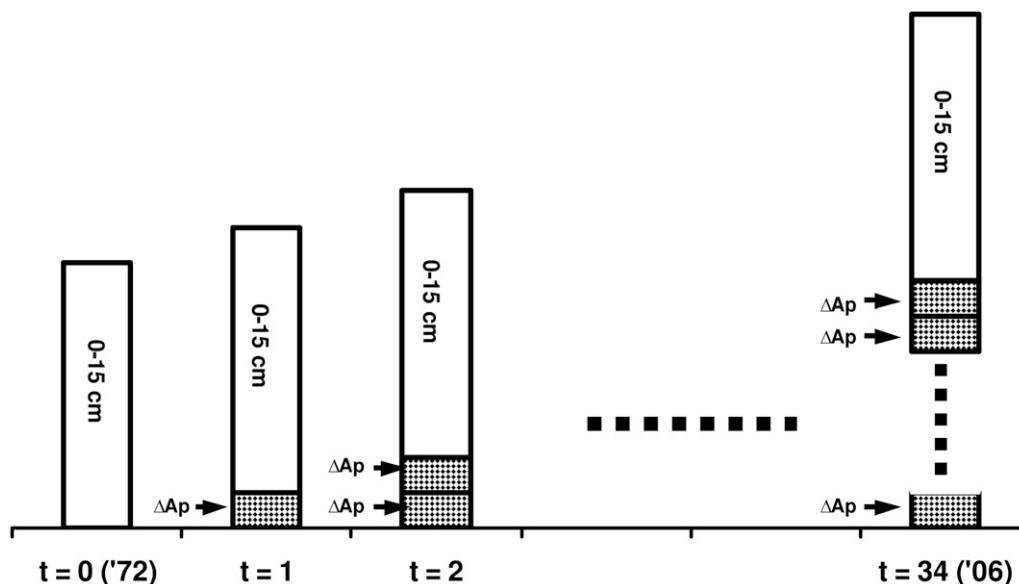


Fig. 1. Hypothetical influence of biosolids application in the previous year on surface soil layer depth (one ΔA_p denotes the annual increase in surface soil layer depth after biosolids application in the previous year).

tion year as the change in rapid fraction and use volatile solids content as a parameter to adjust R. Then,

$$y = [0.339 - (55 - V_s)/55]C_{BS}e^{-0.0205t} + \{1 - [0.339 - (55 - V_s)/55]\}C_{BS}e^{-0.000301t} \quad [6]$$

where y is the remaining biosolids C, V_s is the volatile solids in biosolids, C_{BS} is the C input from biosolids application, and t is time (days).

Because the digester draw-off contained 55% volatile solids, according to Eq. [6], the rapid fraction (0.339) should be completely lost when volatile solids decreased to 36.4%. Subsequently, we treated biosolids in some years having a volatile solids of 36.4% or below with no rapid fraction.

The remaining biosolids C from each of the previous application years was calculated, and a sum was established as total biosolids C remaining in 1985 and 2006.

Estimation of Net Soil Carbon Sequestration

The net soil C sequestration observable in 1985 and 2006 was obtained as the difference between soil C gain and the total biosolids C remaining in 1985 or 2006.

Statistical Analysis

Because the frequency of biosolids applications for three groups was similar up to 1984, we calculated the biosolids C sequestration efficiency in 1985 for comparing the effect of initial soil conditions on the biosolids-induced soil C sequestration. The biosolids C sequestration efficiency (BSCE) in 1985 was calculated as follows:

$$\text{BSCE} = \frac{\text{Net soil C sequestration in 1985}}{\text{cumulative biosolids applied by 1985}} \quad [7]$$

The significance of difference in BSCE between field groups was evaluated using a t test. At the end of the project (2006), we

performed a linear regression procedure to establish a relationship between annual biosolids application and net soil C sequestration that can be used to predict the C sequestration in the biosolids land application. The annual biosolids application or soil C sequestration was calculated through dividing the cumulative biosolids application or total net C sequestration in 2006 by 34. All biosolids applications are reported on a dry-weight basis.

Results and Discussion

Biosolids Organic Matter Content

Table 3 shows variations in chemical characteristics among three types of biosolids applied to the Fulton county fields from 1972 to 2004. The organic matter measured as volatile solids in biosolids decreased in the following order: lagooned liquid > lagooned air-dried > lagooned dewatered. Variations in volatile solids among liquid biosolids of various years reflected the effect of storage time. The liquid biosolids applied at later years had longer storage time, thus having lower volatile solids (Table 3). The low volatile solids in dewatered biosolids were due to the dilution by clay dredged from earthen holding basins and to volatilization and oxidation of organic compounds during the prolonged storage. Similarly, the clay dredged from the lagoon bottom decreased the volatile solids in air-dried biosolids produced in the early 1990s when the storage lagoons at the Stickney and Calumet WRP were not paved. The agitation and air-drying in drying beds, which enhance the organic volatilization and oxidation, are also contributors for low volatile solids in air-dried biosolids. Because most dewatered biosolids and air-dried biosolids from unpaved lagoons were concurrently applied from mid-1980s to early 1990s, the fields received biosolids of relatively low organic matter during that period. The biosolids applied in 1994 and 1995 added extremely low organic matter because the remaining small amount of pre-Part 503 dewatered biosolids was mixed, as allowed, with sand. Variations in organic N, $\text{NH}_4\text{-N}$, Fe, and Al among biosolids types and application years generally fol-

Table 3. Some properties of lagooned biosolids applied to fields for land reclamation at Fulton County, Illinois over a period of 33 yr.

Year	Type of biosolids	Tot solids	Volatile solids	%				C/N	%	
				Org. C‡	Org. N	NH ₄ -N	Total Fe		Total Al	
1972†	liquid	4.0	57.0	33.1	4.01	3.26	4.5	4.56	1.04	
1973†	liquid	4.4	53.9	31.3	3.41	2.50	5.3	3.18	1.06	
1974	liquid	4.1	48.8	28.3	2.20	2.90	5.5	4.40	1.40	
1975	liquid	4.7	46.8	27.1	3.19	2.47	4.8	4.73	1.30	
1976	liquid	4.9	47.6	27.6	2.65	2.10	5.8	5.61	1.57	
1977	liquid	2.9	42.1	24.4	2.70	2.16	5.0	5.06	1.50	
1978	liquid	3.6	43.5	25.2	2.71	2.20	5.1	4.08	1.43	
1979	liquid	3.8	42.4	24.6	2.36	1.68	6.1	4.65	1.23	
1980	liquid	3.7	33.4	19.4	2.82	1.47	4.5	4.27	1.53	
1980	dewatered§	51.8	29.2	16.9	1.08	0.14	13.9	4.19	0.56	
1981	liquid	4.4	44.6	25.9	2.61	1.99	5.6	4.51	1.27	
1981	dewatered§	47.9	22.8	13.2	1.70	0.18	7.0	5.20	1.12	
1982	liquid	3.8	42.8	24.8	2.73	1.71	5.6	4.21	1.40	
1982	dewatered§	63.6	14.6	8.5	0.71	0.02	11.6	1.97	0.51	
1983	liquid	3.1	45.3	26.3	2.67	1.83	5.8	4.14	1.29	
1984	liquid	2.2	38.4	22.3	3.59	1.62	4.3	4.57	1.29	
1984	dewatered§	73.0	14.9	8.6	0.63	0.08	12.2	2.27	0.89	
1985	liquid	6.1	32.5	18.9	2.54	0.96	5.4	4.10	1.11	
1985	dewatered§	66.0	19.7	11.4	0.83	0.05	13.0	2.59	0.56	
1986	dewatered§	64.6	16.9	9.8	0.67	0.08	13.1	2.11	0.78	
1987	dewatered§	30.2	29.5	17.1	1.24	0.52	9.7	1.86	0.42	
1987	air-dried§	66.6	25.9	15.0	1.07	0.11	12.8	2.09	0.78	
1988	dewatered§	25.0	35.4	20.5	1.64	0.54	9.4	1.09	0.30	
1988	air-dried§	69.1	21.1	12.2	0.76	0.01	15.9	1.86	0.66	
1989	dewatered§	34.6	32.7	19.0	1.36	0.59	9.7	4.01	1.29	
1989	air-dried§	70.8	24.4	14.2	0.93	0.32	11.3	2.64	1.07	
1990	dewatered§	40.6	29.1	16.9	1.21	0.32	11.0	3.04	1.29	
1990	air-dried§	69.2	25.3	14.7	0.97	0.18	12.8	2.83	1.11	
1991	dewatered§	37.5	24.8	14.4	1.19	0.34	9.3	5.35	1.31	
1991	air-dried§	68.0	34.9	20.2	1.33	0.41	11.7	2.74	1.37	
1992	dewatered§	23.8	30.8	17.9	1.72	0.70	7.4	4.88	1.50	
1992	air-dried§	66.0	35.6	20.6	1.62	0.43	10.1	2.51	1.55	
1993	no application									
1994	dewatered	ND¶	ND	ND	ND	ND	ND	ND	ND	
1995	dewatered	ND	ND	ND	ND	ND	ND	ND	ND	
1996	no application									
1997	air-dried	70.6	35.7	20.7	1.41	0.32	12.0	3.04	1.14	
1998	air-dried	78.3	38.3	22.2	1.60	0.56	10.3	2.90	1.09	
1999	air-dried	68.9	39.2	22.7	1.71	0.48	10.4	3.15	1.40	
2000	air-dried	68.6	31.1	18.0	1.19	0.14	13.5	2.28	1.11	
2001	air-dried	70.2	35.4	20.5	1.47	0.40	11.0	3.40	1.77	
2002	air-dried	71.3	41.3	24.0	2.00	0.41	9.9	3.74	1.54	
2003	air-dried	70.4	31.0	18.0	1.37	0.23	11.3	ND	ND	
2004	air-dried	65.2	43.7	25.3	1.96	0.41	10.7	ND	ND	
Mean	liquid	4.0	44.2	25.7	2.87	2.06	5.2	4.43	1.31	
	dewatered	46.6	25.0	14.5	1.17	0.25	10.6	3.21	0.88	
	air-dried	69.5	33.1	19.2	1.38	0.31	11.7	2.76	1.22	

† Sampled at the just completion of anaerobic digestion, also called digester drawoff.

‡ Org. C, volatile solids/1.724.

§ Dewatered and air-dried biosolids containing clay inadvertently scraped from the earthen surface of holding basins or unpaved lagoons/drying cells.

¶ ND, not determined.

lowed those for the volatile solids (Table 3). Dewatered and air-dried biosolids had a C/N ratio similar to typical topsoil.

Soil Organic Carbon Concentration Dynamics

Soil organic carbon concentration in control with chemical fertilizer application alone showed a small increase for mine spoil soil (F29) and slight decrease for nonmined soil (F18–2) over the course of the project (Table 4). Application of biosolids rapidly increased the SOC concentrations up to 1985 in fields of all

three groups. The SOC concentration in the fields of all groups started to peak in the mid-1980s and maintained that level for nearly 10 yr in Groups I and II fields where biosolids were applied every year. The SOC in Group III started to decline after peaking at 2 to 3 yr because the application of biosolids in most fields of this group was ceased at that time. At the highest loading rates (1400–1700 Mg ha⁻¹ in 34 yr or about 42–49 Mg ha⁻¹ yr⁻¹) in Group I and II fields, the SOC was maintained between 6.5 to 7% in the 0- to 15-cm depth during the 3 yr (1993–1995).

Table 4. Dynamics of soil organic carbon concentration (%) at 0- to 15-cm depth along with biosolids application in fields of three groups at Fulton County land reclamation site (listed from low to high rates of cumulative biosolids application in each group).

Field no.	'72	'73	'74	'75	'76	'77	'78	'79	'80	'81	'82	'83	'84	'85	'86	'87	'88	'89	'90	'91	'92	'93	'94	'95	'96	'97	'98	'99	'00	'01	'02	'03	'04	'05	'06		
Group I: mine spoil soil																																					
F29(Fert CL)	ND	ND	ND	ND	0.62	ND	0.84	0.67	0.71	0.94	0.89	ND	0.87	0.87	1.04	1.08	0.90	1.07	0.97	1.04	1.08	0.92	1.03	0.96	0.89	0.76	1.08	0.94	0.81	0.86	0.96	0.91	0.90	1.12	0.96		
F30	ND	ND	0.67	0.56	1.12	1.70	1.99	2.59	3.27	3.10	3.60	3.66	4.02	4.56	4.24	4.23	4.28	4.44	4.24	4.24	3.98	4.02	3.98	4.02	3.87	3.95	3.52	3.62	3.59	3.32	3.88	3.68	3.95	3.51	4.13	4.11	
F31	ND	ND	ND	0.73	ND	0.98	1.96	1.99	1.98	3.76	4.34	4.91	3.63	3.90	4.27	3.08	3.66	3.30	3.66	3.30	3.85	3.71	3.58	3.28	2.99	3.38	3.42	3.39	2.53	2.38	2.04	3.29	3.24	3.02	3.68	3.49	
F32	ND	ND	1.28	1.45	2.10	2.11	3.39	4.01	4.51	5.36	5.88	6.95	6.76	6.93	6.59	6.85	6.32	6.20	6.25	6.26	5.41	4.77	5.46	5.62	5.68	5.10	5.16	4.85	5.09	5.39	4.74	5.12	4.46	4.81	4.55		
F33	ND	ND	ND	0.98	1.87	2.66	3.36	3.37	3.29	4.29	5.83	5.37	5.05	5.61	5.56	5.60	4.95	5.34	5.02	4.63	4.55	5.14	4.81	4.32	4.31	3.78	4.81	3.92	3.95	3.71	3.78	4.52	3.85	4.09	3.64		
F34	ND	ND	0.66	0.69	1.38	2.34	3.10	3.35	4.06	4.63	5.09	5.39	5.96	6.51	5.77	5.56	6.00	5.83	5.74	5.49	5.49	5.37	5.78	5.38	4.76	4.93	4.38	4.71	4.38	4.10	4.73	4.47	4.58	4.07			
F26	ND	ND	1.15	1.16	1.59	2.41	3.59	3.13	4.74	5.65	5.40	5.68	5.14	5.32	5.63	5.53	6.08	5.82	5.41	5.01	5.03	4.64	4.67	4.73	4.15	4.37	4.00	4.71	4.33	4.29	3.85	4.29	3.88	3.67			
F13	ND	ND	0.34	0.69	0.97	1.62	2.58	2.85	4.05	4.60	5.18	5.56	5.61	5.95	5.51	5.72	5.81	5.35	5.37	5.76	5.60	4.67	5.63	5.31	4.96	4.11	4.45	4.21	3.96	4.27	5.34	4.67	4.04	4.99	4.55		
F12	ND	ND	0.52	0.59	0.99	1.86	2.98	3.67	3.61	4.12	4.11	4.15	4.30	4.73	4.76	4.98	5.13	4.79	4.73	4.70	4.82	4.10	4.99	4.06	4.13	3.65	3.47	3.54	3.74	4.61	4.74	4.49	3.70	3.89	3.35		
F33	ND	ND	0.57	0.37	0.99	1.99	2.37	3.15	3.77	4.26	4.14	4.90	4.85	5.00	5.74	5.07	5.67	6.09	6.98	6.96	6.80	6.24	6.31	5.11	4.80	5.44	5.27	5.25	6.19	5.77	5.49	5.45	5.09	5.09	5.09		
F8	ND	ND	0.22	0.37	0.95	1.97	2.22	2.82	3.18	3.31	4.22	4.10	4.00	4.57	5.14	4.67	4.30	4.68	5.70	5.22	6.29	5.05	5.27	4.49	4.27	3.90	4.20	4.09	4.23	3.90	4.34	4.71	4.57	4.24	4.14		
F30	ND	ND	0.53	0.84	1.06	1.89	3.57	3.57	2.94	4.73	4.98	5.26	5.14	4.85	4.69	4.94	5.30	5.02	4.90	5.16	5.36	4.77	5.11	4.57	5.05	4.47	4.28	4.74	4.62	4.75	4.55	4.15	4.08	3.88	3.67		
F11	ND	ND	0.73	1.15	1.48	3.17	3.51	4.20	5.09	5.14	5.41	5.23	5.28	5.42	4.43	4.74	4.34	5.22	5.56	5.82	5.58	5.09	5.67	5.54	5.37	4.77	4.33	4.40	4.39	4.58	4.11	4.76	4.13	4.23	4.35		
F14	ND	ND	0.67	0.80	0.72	1.41	2.13	2.68	2.87	4.13	4.37	4.21	4.19	4.95	4.87	4.80	4.44	4.24	5.25	4.67	5.11	4.94	4.91	4.85	4.58	3.87	4.04	3.74	3.74	4.41	4.52	3.93	4.42	4.13			
F4	ND	ND	0.63	1.13	1.26	2.78	3.99	3.74	4.54	4.63	5.25	5.00	5.32	5.90	6.28	5.82	6.00	5.84	5.96	5.71	5.58	4.59	5.23	5.03	4.62	4.23	4.35	5.43	5.23	4.95	4.47	5.44	4.84	5.33	5.01		
F15	ND	ND	0.77	0.70	1.22	2.02	3.54	3.10	4.50	4.55	4.79	5.12	4.31	4.96	4.44	4.64	5.16	5.08	4.75	5.69	4.23	4.89	5.21	4.86	4.44	3.99	3.67	4.05	3.44	3.57	4.01	4.45	4.05	4.50	4.07		
F5	ND	ND	0.38	0.89	1.14	2.29	2.52	3.30	3.56	3.46	3.87	3.99	4.10	3.73	4.08	4.24	4.32	4.20	4.49	5.02	5.04	5.38	5.26	5.16	5.28	4.37	5.00	4.87	5.27	5.10	5.65	5.81	5.25	4.97			
F7	ND	ND	0.31	0.59	1.20	0.98	2.29	2.66	2.93	3.28	2.97	2.84	3.09	3.02	3.22	2.80	3.31	3.48	3.25	4.20	3.78	3.34	3.42	3.30	3.07	3.28	4.07	3.57	3.30	3.07	3.32	3.53	3.60	3.41			
F3	0.80	ND	0.86	0.96	1.23	2.24	3.09	3.80	4.15	4.57	4.79	4.37	5.29	5.60	5.54	5.54	5.54	5.54	5.67	5.67	6.32	6.96	6.00	6.75	6.59	4.96	4.87	4.75	5.32	5.76	6.37	6.39	5.35	6.28	5.88		
F1	ND	ND	0.69	0.75	1.30	2.31	2.82	3.62	3.71	4.31	4.48	5.35	5.36	5.80	5.56	5.75	5.29	5.44	5.01	5.86	6.83	6.45	6.12	5.79	6.15	5.33	5.50	5.42	5.31	4.84	5.22	5.22	4.99	5.37	5.26		
F2	ND	ND	0.55	0.95	1.28	2.74	3.39	3.73	4.49	4.90	4.20	4.86	4.24	5.06	4.86	4.72	4.35	4.69	5.60	5.39	6.26	5.11	5.52	5.20	4.01	4.83	5.12	5.86	4.73	4.97	4.63	5.80	5.11	5.12	5.18		
Group II: "fine" mine spoil soil (mine spoil + pre-mined soil)																																					
F18-2	ND	ND	ND	ND	2.37	ND	2.45	ND	2.61	1.98	1.90	ND	2.26	2.15	2.27	2.00	1.74	1.93	ND	1.93	ND	2.33	ND	2.39	2.04	1.77	1.99	ND	1.91	ND	1.79	2.21	ND	2.22	2.24		
(Fert CL)†	ND	ND	1.94	ND	2.32	2.53	2.70	3.45	3.85	4.14	4.65	5.36	5.75	6.27	5.19	5.45	5.27	4.82	5.00	4.47	5.06	4.22	4.40	4.53	4.48	3.88	3.65	4.01	3.74	3.40	3.94	3.81	3.65	3.66			
F40	ND	ND	0.74	ND	0.77	1.14	1.76	2.76	4.20	4.57	4.49	4.99	5.67	5.11	5.58	4.76	4.37	5.20	4.75	4.61	3.98	4.35	4.54	4.56	3.77	3.96	3.68	3.67	3.26	3.70	3.30	3.62	4.26	3.97			
F44	ND	ND	0.89	ND	0.81	1.34	1.72	1.96	3.59	3.69	3.84	4.59	4.29	4.48	4.61	4.30	4.07	4.09	4.18	4.20	3.96	3.75	4.02	4.18	3.67	3.63	3.59	4.00	4.04	3.85	3.59	4.18	4.16	3.78			
F45	ND	ND	1.12	ND	1.34	1.95	2.33	3.20	3.82	4.19	4.36	5.13	4.84	5.34	4.89	5.38	4.83	3.91	5.01	4.94	4.21	4.61	3.63	4.29	3.71	3.83	3.97	3.65	3.88	3.50	4.04	4.58	3.95	3.96			
F42	ND	ND	1.12	ND	1.33	2.14	2.56	2.22	3.11	3.78	4.20	4.55	4.22	3.62	5.00	4.63	3.80	4.02	3.71	3.70	4.48	3.33	3.50	3.59	3.54	5.07	4.78	4.15	4.55	5.45	5.31	4.53	4.66	4.90			
F43	ND	ND	0.95	ND	1.67	2.08	2.51	3.50	4.01	4.18	4.14	4.31	5.25	5.27	5.15	4.78	4.16	3.95	4.44	4.25	4.22	3.93	3.90	3.92	3.52	4.52	4.61	3.91	4.13	4.38	3.96	4.07	5.09	4.11			
F47	ND	ND	0.80	ND	0.96	1.90	2.53	3.65	4.69	4.53	5.45	5.68	6.76	6.96	6.56	6.23	5.61	5.92	5.88	6.79	6.85	6.98	6.85	6.85	6.72	5.58	5.81	5.38	5.19	4.98	5.74	5.62	5.77	5.95	5.09		
F9	ND	0.86	0.65	1.12	1.27	1.80	2.39	3.43	3.53	3.92	4.10	4.57	4.52	5.14	5.57	4.92	5.27	5.26	6.33	5.50	6.86	5.85	5.81	5.23	5.08	4.62	4.25	4.72	5.17	4.43	4.76	5.08	5.53	4.98	4.58		
F17	ND	ND	1.05	ND	1.24	1.13	1.62	3.14	4.42	4.92	5.63	5.08	6.11	6.03	6.01	6.94	6.29	6.02	7.04	6.54	6.61	6.86	5.48	5.70	5.64	5.83	5.71	5.94	6.17	5.32	6.50	5.76	5.76	5.57			
Group III: nonmined soil																																					
F18-2	ND	ND	ND	ND	2.37	ND	2.45	ND	2.61	1.98	1.90	ND	2.26	2.15	2.27	2.00	1.74	1.93	ND	1.93	ND	2.33	ND	2.39	2.04	1.77	1.99	ND	1.91	ND	1.79	2.21	ND	2.22	2.24		
(Fert CL)†	ND	1.19	1.23	1.01	1.24	1.96	2.60	3.76	4.20	4.30	4.78	5.75	4.76	5.49	5.05	5.73	4.91	5.00	4.36	4.75	4.33	4.40	4.41	4.31	4.36	4.16	3.75	3.94	3.34	3.47	3.47	3.29	3.18	3.03	2.93		
F22	ND	1.06	ND	1.50	1.30	2.50	3.12	3.68	3.82	4.59	4.87	4.25	5.67	5.24	4.85	3.31	4.25	3.19	4.23	2.75	3.46	3.80															

The SOC did not continue to rise in the late 1980s and early 1990s; this was probably due to low organic matter in dewatered biosolids and air-dried biosolids from unpaved storage lagoons concurrently applied during those years. The SOC showed some declines in the late 1990s because no biosolids were applied in 1993 and 1996, and in 1994 and 1995 the biosolids were diluted by sand. In some fields of Group III where biosolids application was ceased 22 yr ago, the SOC in 2006 was still well above the initial levels. The loss of SOC after the cessation of biosolids application followed the first-order decay model. The decomposition rate, k , ranged from 0.0225 to 0.0339 yr⁻¹ or 2.2 to 3.3% loss annually (on average, 0.0276 yr⁻¹ or 2.7%).

Topsoil having a stable organic matter of 5% or organic carbon at 3% is usually considered productive in the region because this level of organic matter usually gives the soil desirable chemical, biological, and physical properties. Table 4 shows that the SOC could be maintained above 3% with the application of biosolids of 23.6 Mg ha⁻¹ yr⁻¹ in a Group III field (non-mined F37). Because this biosolids loading rate is equivalent to the average rate currently used in the MWRDGC's farmland application program where biosolids are applied as N fertilizer, the continuous application of biosolids at the rate to meet crop N requirement would maintain the SOC at a level of productive soil. Brown and Leonard (2004) reported that the SOC was greater in the biosolids plots with 10 yr of continuous low-rate application than in N-fertilizer plots.

Farmyard manure is the most recognized among the commonly used organic amendments in its ability to maintain and build SOM (Paustian et al., 1997). Parat et al. (2005) reported that biosolids restored the SOC to a level as high as farmyard manure on a French sandy soil. Gerzabek et al. (2001) reported that the increase in SOC of a Swedish soil was higher in biosolids than in animal manure. The intensive microbial decay of the organic matter during anaerobic digestion leaves the biosolids as a relatively recalcitrant residue, and lagooning further reduced the decomposability of the biosolids (Gilmour et al., 2003). This makes biosolids become an ideal organic amendment to build SOM.

The ultimate goal of the MWRDGC's land reclamation is to bring the SOC to the level of Midwest prairie, apart from improving soil physical properties. Reliable estimates of SOC concentration in pre-cultivation Illinois prairie soil are not available because SOC monitoring began some time after the establishment of cultivation experiments (Darmody and Peck, 1997). Based on the first available SOC of 2.4% C in 1904 in the continuous corn plots established in 1876 by University of Illinois as the oldest agronomic research fields in the USA and the oldest continuous corn plots in the world (Darmody and Peck, 1997) and assuming that the SOC loss after cultivation was 2.7% per year as observed in the region, the dark prairie soils (Flanagan silt loam: fine montmorillonitic, mesic, Aquic Argiudoll) probably contained 5.3% SOC before cultivation in 1876. The prairie soil at Nashua (Iowa), 300 km northwest of Fulton County, has a SOC of 6.61% in the 0- to 15-cm depth (Russell et al., 2005). Two fields (F3 and F17) with a mean annual loading rate of 42 Mg ha⁻¹ showed an SOC level of between 5.3 and 6.6% in 2006 (Table 4). These data indicate that the land reclamation using

biosolids was able to restore the SOC of stripmined and degraded soils to a level similar to the prairie soils in a few decades.

Soil Carbon Sequestration

Application of chemical fertilizer alone over three decades led to a soil C gain of 5.7 Mg ha⁻¹ in mine spoil soil and a loss of 2.4 Mg ha⁻¹ in nonmined soil (Table 5). Application of biosolids during the same period resulted in a remarkable soil C gain: 74.4 to 166 Mg ha⁻¹ in Group I fields, 37.1 to 139 Mg ha⁻¹ in Group II fields, and 24.8 to 107 Mg ha⁻¹ in Group III fields (Table 5). After subtracting the biosolids C remaining in 2006 (ranging from 17.5 to 62.6 Mg ha⁻¹ in Group I fields, 6.4 to 61.4 Mg ha⁻¹ in Group II fields, and 5.0 to 35.7 Mg ha⁻¹ in Group III fields), the net soil C sequestration due to biosolids application was still remarkably high (ranging from 37.5 to 104 Mg C ha⁻¹ in Group I fields, 28.2 to 86.9 Mg C ha⁻¹ in Groups II fields, and 18.2 to 83.5 Mg C ha⁻¹ in Group III fields) (Table 5). The mean net soil C sequestration in the biosolids-amended fields in this study was 1.73 Mg ha⁻¹ yr⁻¹ (range, 0.54– 3.05 Mg ha⁻¹ yr⁻¹).

Data from Varvel (2006) indicate that unless a cover crop such as clover was included, both mono- and multiple-cropping, even with adequate N fertilizer, had a negative soil C balance from 1984 to 2002 in Nebraska. West and Post (2002) analyzed published and unpublished data of C sequestration rates of 67 long-term agricultural experiments with 276 paired treatments and found that a change from conventional tillage to no-tillage could sequester 0.57 Mg C ha⁻¹ yr⁻¹ in soil. They also noted that such soil C sequestration peaked in 5 and 10 yr and declined to zero in 15 to 20 yr. Paustian et al. (2002) predicted Iowa agricultural soils could gain C of 0.25 Mg ha⁻¹ yr⁻¹ by changing from intensive to moderate-till practices based on the Century SOM model. The high soil C sequestration in the biosolids-amended fields observed in this study is due to several important factors.

Biosolids application at the land reclamation fields increased soil microbial biomass (Tian et al., unpublished data). As a precursor to more stable fractions of SOM (Parton et al., 1987), increase in the size of microbial biomass leads to the increase in SOM (Powlson et al., 1987). Although the formation of SOM through microbial biomass can be the re-distribution of original SOC and biosolids C rather than C sequestration, some of the microorganisms are autotrophic (e.g., nitrifiers, which are able to assimilate CO₂ as a C source) and contribute to C sequestration. Some heterotrophic organisms require CO₂ for the formation of biomass. The improvement in microbial biomass has been one of the reasons for C sequestration in no- and minimal tillage (Wright et al., 2005).

Biosolids contain appreciable amounts of Fe and Al (Table 3). Measurement at another experiment with nonmined soil within the land reclamation site indicates that application of biosolids in 2005 significantly increased the soil oxalate-extractable Fe and Al in 2006. The oxalate-extractable Fe and Al are amorphous Fe and Al oxides. These noncrystallines are the most reactive Fe and Al oxides due to their small size and consequently high surface area. Buurman et al. (2007) found that allophane could contribute to the increase in SOM through incorporating decomposition prod-

Table 5. Soil carbon sequestration in fields of three groups after 34-yr land reclamation with biosolids at Fulton County, Illinois (listed from low to high rates of cumulative biosolids application in each group).

Field no.	Initial surface soil layer C stock	2006 surface soil layer C stock	Soil C gain	Cum biosolids C applied‡	Biosolids C remaining	Net soil C sequestration
Mg ha ⁻¹						
Group I: mine spoil soil						
F29 (Fert CL)	15.0	20.6	5.7	0.3	0	5.7
F32	16.2	106	90.1	148	40.4	49.8
F39	17.6	92.1	74.4	154	36.9	37.5
F25	27.5	117	89.7	172	17.5	72.2
F27	22.5	97.2	74.7	196	23.8	50.9
F28	15.9	111	95.0	216	25.6	69.5
F26	25.2	102	76.6	217	19.4	57.2
F13	8.2	142	133	231	48.0	85.5
F12	12.6	110	97.6	204	24.8	72.8
F33	13.8	145	131	246	48.3	82.5
F8	5.3	126	121	225	33.4	87.7
F30	12.8	108	95.7	257	31.8	63.9
F11	17.6	137	119	251	26.2	93.3
F14	16.2	131	115	229	48.5	66.6
F4	15.2	151	136	274	45.0	90.6
F15	18.6	128	109	268	51.8	57.2
F5	9.2	149	140	277	47.8	91.8
F7	7.7	108	100	263	42.0	57.8
F3	19.3	186	166	295	62.6	104
F1	16.7	170	154	305	56.7	97.1
F2	13.3	165	152	341	56.7	95.1
Group II: "fine" mine spoil soil (mine spoil + pre-mined soil)						
F18-2 (Fert CL)†	42.1	39.6	-2.4	0.3	0	-2.4
F40	34.6	71.7	37.1	124	6.4	30.8
F44	14.3	90.2	75.9	190	40.4	35.5
F45	17.2	87.0	69.8	185	35.8	34.0
F42	21.5	88.5	67.0	189	25.1	41.9
F41	21.7	115	93.7	228	61.4	32.3
F43	18.4	99.1	80.7	228	52.5	28.2
F47	15.5	126	111	260	51.6	59.4
F9	16.6	135	118	240	40.3	77.8
F17	20.3	160	139	292	52.5	86.9
Group III: nonmined soil						
F18-2 (Fert CL)	42.1	39.6	-2.4	0.3	0	-2.4
F22	23.0	63.8	40.8	104	5.0	35.8
F23	20.5	56.2	35.7	108	5.4	30.3
F20	38.8	66.8	28.0	131	6.4	21.5
F31	9.3	47.7	38.4	138	6.3	32.0
F34	9.3	57.0	47.7	130	6.5	41.2
F21	19.4	54.5	35.1	136	6.3	28.8
F19	28.4	53.2	24.8	132	6.6	18.2
F37	14.3	79.5	65.2	190	19.8	45.4
F35	18.1	102	84.0	229	35.7	48.3
F10	9.5	116	106	238	22.9	83.5
F36	13.0	101	87.6	249	35.3	52.3
F16	13.6	104	90.7	214	23.9	66.8

† Nonmined soil, but similar to Group II.

‡ Calculated assuming biosolids organic C = volatile solids/1.724.

ucts and microbial SOM in very fine aggregates. Mikutta et al. (2006) noted the interaction of poorly crystalline minerals with SOM promoted SOM stabilization. Kaiser et al. (2002) pointed out that the intimate association of organic matter with secondary hydrous Fe and Al phases against biological degradation could be the chief reason for survival of organic matter in subsoils of two forest soils in Germany. Huggins et al. (1998) measured plant biomass C input to soil in various rotation systems in Minnesota from 1981 to 1990. They found that a corn-soybean rotation could provide soil a C input of 5.4 Mg ha⁻¹ yr⁻¹ via above- and

below-ground biomass of corn and soybean. Collins et al. (1999) estimated that the total biomass C input ranged from 6.1 to 7.6 Mg C ha⁻¹ in continuous corn within the Corn Belt of the USA. However, the humification rate constant for added biomass C was only 0.16 yr⁻¹ for corn and 0.11 yr⁻¹ for soybean (Huggins et al., 1998). Therefore, potentials exist for more biomass C to be transformed to SOM. The Fe and Al from biosolids might have acted as an agent that increased that humification rates of corn and soybean biomass C in this study. Improvement in soil aggregation by biosolids also increased the stability of SOM (Tiessen et al.,

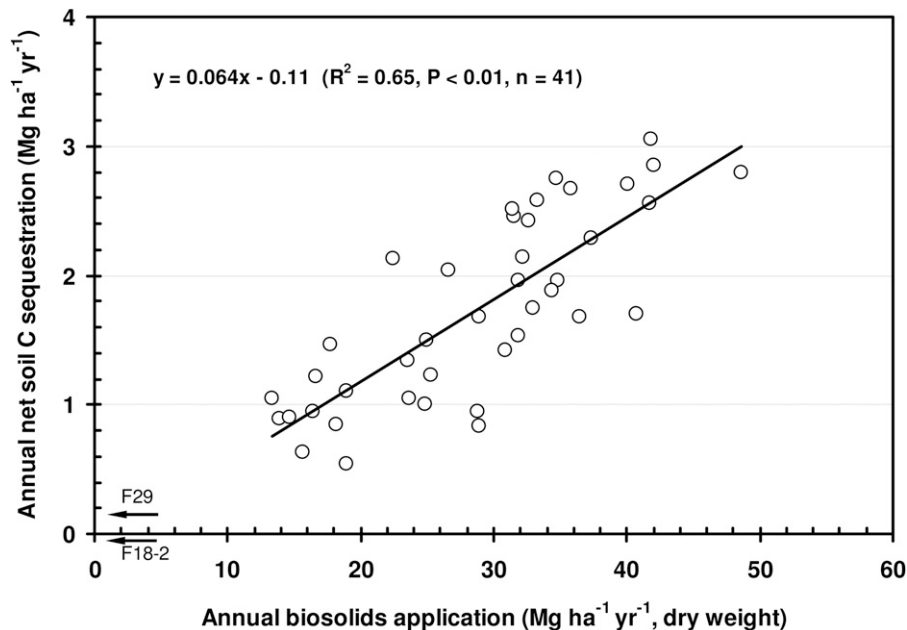


Fig. 2. Correlation between annual net soil C sequestration and biosolids application in fields of all three groups at Fulton County land reclamation site. Data are derived from Tables 1 and 5. The arrow points to the rate of soil C sequestration in fertilizer control field 29 (F29) and field 18–2 (F18–2).

1984; Six et al., 2002). Further study by analysis of ^{13}C is being envisioned so that a more firm conclusion regarding biosolids applications and C sequestration can be drawn.

Regression analysis revealed that soil C sequestration was positively correlated to biosolids loading rate (Fig. 2). Based on the equation in Fig. 2, annual application of biosolids at a level meeting corn N requirement, normally $22.4 \text{ Mg ha}^{-1} \text{ yr}^{-1}$, would lead to a soil C sequestration of 1.3 Mg C ha^{-1} .

Until 1984, the frequency of biosolids application was similar for the three groups (see Table 2). Therefore, by comparing the soil C sequestration efficiencies between three groups in 1985, we can evaluate the effect of initial soil conditions on soil C sequestration efficiency in the biosolids land reclamation. The results of this evaluation showed that biosolids soil C sequestration efficiency (1985) was greater in Group I than in Group II or III (Table 6). Soils in Group I were more degraded than those in Group II and III because Group II fields got back the pre-mined soils and Group III fields were not mined. The average initial SOC was 0.66% for Group I fields, 1.05% for Group II fields, and 0.92% for Group III fields. Tian (1998) reported that soil degradation reduced the decomposition of organic matter because it caused the loss of soil biota and deterioration of soil physical and chemical properties, resulting in longer residence of organic matter. It is possible that higher degradation in Group I soils led to lower SOM decomposition, thus allowing more SOM to be retained as compared with other groups.

There are some limitations in this study. After long-term biosolids application, slight changes are expected for SOC content in subsoil, which may affect our C sequestration estimation approach. The soil bulk was estimated, which could be a potential error for the soil C stock; however, we believe our bulk density estimates should be close to reality as verified from our 2005

samples. Also, it might not be feasible to monitor soil bulk density for a period of three decades on the 1000-ha research fields. We are confident that our approach to estimate soil C stock was the best method considering these field situations. To our knowledge, there is no better method available for estimating the SOC stock in a situation in which surface soil layer depth and mass changes under management. Under such conditions, the equal soil weight method (Ellert and Bettany, 1995; Ellert et al., 2001), commonly used for estimating SOC stock, cannot be applied. Recently, Chang et al. (2007) proposed to measure the change in elevation to calibrate soil C stock for the situation, in which organic additions modify the soil depth, but it is not possible to do so annually because the change in soil depth over 1 yr is too small to measure. Also, the approach for sampling by the genetic horizon is not adequate for this large-scale experiment. Our continuous sampling helped to obtain highly reliable C sequestration from this project. Another major error in our soil C sequestration estimation comes from biosolids C remaining. Nevertheless, our estimates should not deviate much from the real measurements because we have considered many factors, including the site soil moisture and temperature, in establishing a biosolids decomposition equation. Our approach of estimating biosolids C remaining for obtaining the net soil C sequestration is conservative because our biosolids decomposition estimates were low compared with the majority of data in the literature.

Conclusions

The high SOC improvement and soil C sequestration in biosolids-amended fields of a large-scale, long-term land reclamation project can allow us to draw some important conclusions for searching approaches for land reclamation and the management of biosolids. Biosolids are an effective material in increasing SOM

Table 6. Efficiency of soil carbon sequestration by biosolids application in different land reclamation fields as affected by field group (listed from low to high rates of cumulative biosolids application in each group).

Field no.	Cumulative biosolids applied (72–84) Mg ha ⁻¹	Net soil C	C sequestration efficiency Net soil C sequestration/ cumulative biosolids mass
		sequestration (1985)	
Group I: mine spoil soil (MS)			
F32	388	54.1	0.139
F39	442	24.6	0.056
F25	569	70.8	0.124
F27	616	38.7	0.063
F28	707	58.1	0.082
F26	704	29.8	0.042
F13	535	84.7	0.158
F12	642	54.0	0.084
F33	635	41.6	0.066
F8	485	59.1	0.122
F30	722	40.0	0.055
F11	662	64.2	0.097
F14	562	40.8	0.073
F4	634	69.9	0.110
F15	704	44.4	0.063
F5	556	32.3	0.058
F7	520	19.4	0.037
F3	496	57.4	0.116
F1	630	63.4	0.101
F2	662	54.5	0.082
Mean			0.086
Group II: "fine" Mine spoil soil (Mine spoil + pre-mined soil) (fMS)			
F40	497	17.2	0.034
F44	579	33.8	0.058
F45	481	13.7	0.028
F42	622	10.5	0.017
F41	505	6.5	0.013
F43	577	25.5	0.044
F47	664	45.9	0.069
F9	466	37.6	0.081
F17	530	49.4	0.093
Mean			0.049
Group III: nonmined soil (NM)			
F22	455	46.2	0.102
F23	473	45.9	0.097
F20	531	11.8	0.022
F31	557	15.4	0.028
F34	566	39.4	0.070
F21	618	27.5	0.045
F19	644	16.1	0.025
F37	564	23.3	0.041
F35	546	39.3	0.072
F10	615	54.3	0.088
F36	578	38.6	0.067
F16	639	42.8	0.067
Mean			0.060
			Probability of t test
MS vs. fMS			0.006
MS vs. NM			0.024
fMS vs. NM			NS

in strip-mined land and low-productivity soil. The biosolids application could turn the Midwest corn–soybean system soils for C-neutral to C-sink. Biosolids application is a good option that can return the Midwest soil to prairie SOM condition.

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