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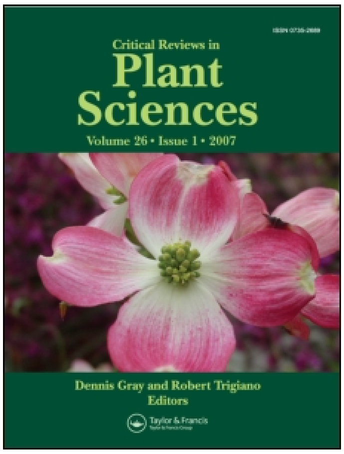

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Mechanisms of Carbon Sequestration in Soil Aggregates

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Table of Contents

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Soil and crop management practices have a profound impact on carbon (C) sequestration, but the mechanisms of interaction between soil structure and soil organic C (SOC) dynamics are not well understood. Understanding how an aggregate stores and protects SOC is essential to developing proper management practices to enhance SOC sequestration. The objectives of this article are to: (1) describe the importance of plants and soil functions on SOC sequestration, (2) review the mechanisms of SOC sequestration within aggregates under different vegetation and soil management practices, (3) explain methods of assessing distribution of SOC within aggregates, and (4) identify knowledge gaps with regards to SOC and soil structural dynamics. The quality and quantity of plant residues define the amount of organic matter and thus the SOC pool in aggregates. The nature of plant debris (C:N ratio, lignin content, and phenolic compound content) affects the rate of SOC sequestration. Mechanisms of interaction of aggregate dynamics with SOC are complex and embrace a range of spatial and temporal processes within macro- (>250 μm e.c.d.) and microaggregates (<250 μm e.c.d.). A relevant mechanism for SOC sequestration within aggregates is the confinement of plant debris in the core of the microaggregates. The C-rich young plant residues form and stabilize macroaggregates, whereas the old organic C is occluded in the microaggregates. Interactions of clay minerals with C rich humic compounds in correlation with clay mineralogy determine the protection and storage of SOC. Principal techniques used to assess the C distribution in aggregates include the determination of total organic C in different aggregate size fractions, isotopic methods to assess the turnover and storage of organic C in aggregates, and computed tomography and X-ray scattering to determine the internal porosity and inter-aggregate attributes. The literature is replete with studies on soil and crop management influences on total organic C and soil aggregation. However, research reports on the interactions of SOC within aggregates for C sequestration are scanty. Questions still remain on how SOC interacts physically and chemically with aggregates, and research is needed to understand the mechanisms responsible for the dynamics of aggregate formation and stability in relation to C sequestration.

Keywords soil structure, organic binding agents, soil organic carbon sequestration, tillage, residence time, macroaggregates, microaggregates, isotopic methods, clay minerals

I. INTRODUCTION

The architectural organization of sand, silt, and clay by organic compounds and inorganic cementing agents creates aggregates. The degree of adherence of these primary particles as a function of organic matter and clay mineralogy determines whether the aggregates are stable or unstable. The organization of these aggregates and the pore space among them is called soil structure (Tisdall, 1996). Soil structure is a dynamic fabric that controls the movement of water and air in the soil and affects the strength and stability of the soil and the overall soil quality for plant growth and storage of soil organic carbon (SOC). Soil structure integrates all mechanisms of interactions among physical, chemical, and biological properties of the soil.

Soil structure and soil organic matter (SOM) are two of the most dynamic properties that are extremely sensitive to crop and soil management. The SOM is a vital soil attribute, enhancing soil quality and its productivity. It is related to innumerable soil functions including fertility (Bauer and Black, 1994; Reeves, 1997), erosion (Lal *et al.*, 1997; Lal, 2003a), tilth (Singh *et al.*, 1992), and water retention and transmission characteristics (Radke and Berry, 1993). Most importantly, SOM is closely related to SOC dynamics in the soils because it constitutes the largest terrestrial reservoir of SOC. Interactions between soil structure and SOM determines the magnitude of the SOC pool. The SOM is a dynamic factor responsible for soil structure development (Beare *et al.*, 1994a). Stable soil structure, in turn, stores and prevents SOM from rapid decomposition. Although often used interchangeably, SOM differs from SOC, and it contains about 55% SOC and 45% other essential elements.

Literature is replete with information on the effects of land management and living biomass on the C cycle and sequestration (Kimble and Stewart, 1995; Lal *et al.*, 1998). There are also numerous studies with regard to the importance of SOM to soil fertility, soil erosion, and plant growth (Schimel, 1995; Gale and Cambardella, 2000). These studies have improved our understanding of the interrelationships between soil biological properties and SOC, and the implications of crop and soil management to C sequestration (Angers, 1992; Six *et al.*, 1999a), but information on the fundamental physical and chemical processes influencing formation and stabilization of aggregates in relation to SOC sequestration is limited. Although it is well known that soil can be a major sink of atmospheric CO_2 (Lal *et al.*, 1997; Follet, 2001; Lal, 2004), mechanisms of interaction between soil structure and SOC dynamics are not well understood (Oades and Waters, 1991; Sainju *et al.*, 2003). Soil aggregation being the nucleus of all mechanisms of C sequestration, understanding how an aggregate stores and interacts with SOC is essential to developing management strategies toward the enhancement of C sequestration at regional and global scales.

The objectives of this article are to: (1) describe the importance of plants and soil functions on SOC sequestration, (2) review the mechanisms of SOC sequestration within aggregates under different soil and crop management practices, (3) explain methods of assessing the distribution of SOC within aggregates, and (4) identify knowledge gaps with regard to SOC and structural dynamics. These objectives are based on the hypothesis that a strong interdependence of aggregation and SOC governs C sequestration. This article builds on previous reviews (Tisdall, 1996; Kay, 1998 and others) and presents a state-of-the-knowledge compendium on soil structure in relation to C sequestration, which is crucial to environmental quality and agricultural sustainability.

A. Importance of Plant and Soil Organic Matter in Aggregation

The amount of plant residues and the degree of SOM decomposition are vital factors to the formation and stabilization of aggregate structure for SOC sequestration (Haynes and Beare, 1996). Soil fauna and flora including plant and animal residues are intrinsically associated with soil aggregation. There is a trade-off between plant growth and aggregation. Soil structure moderates soil and plant functions and is the framework for water, air, and nutrient flow to plants. Plants, in turn, furnish the soil with fresh residues and roots for aggregate structure development. The nature and properties of aggregates are thus determined by the quantity and quality of coarse residues and humic compounds and by the degree of their interaction with soil particles (Jastrow *et al.*, 1998). Plant roots and residues are the primary organic skeleton to enmesh the soil particles together and build aggregates. As the SOM decomposition proceeds, organic particles associate with soil matrix to form macroaggregates ($>$250 um) and microaggregates ($<$250 um) and sequester SOC. The persistent organic fraction is by far critically more important than coarse organic residues to long-term SOC sequestration. Interactions between organic residues and aggregates depend on the origin of residues. Stable macroaggregates have significantly higher root-derived aggregate ^{14}C and particulate organic matter (POM) than unstable macroaggregates but not in surface-derived aggregates ^{14}C and POM, indicating that root exudations associated with microbially derived organic substances stabilize macroaggregates more than inputs of C through surface residues (Gale *et al.*, 2000a,b).

Microbial processes acting on plant residues control the aggregation of soil particles (Watts *et al.*, 2001). They are responsible for the generation of organic-derived enmeshing fabric for aggregation by decomposing coarse and fine organic residues. Plant residues, as a food source, are the medium for the biological activity of soil organisms whose action defines the turnover and formation of aggregates. Understanding of aggregate dynamics in relation to SOC storage requires the integration of concepts about the dynamic functions of plants, soil organisms, and the soil (Angers and Giroux, 1996).

The role of plants and SOM on soil aggregation is of paramount importance to SOC sequestration. Plants are crucial to the atmosphere-biosphere-soil interactions as they control soil C fluxes to the atmosphere, as soils sequester CO_2 through plants. Nevertheless, the rate of SOM decomposition is a function of the quality of plant residue and its accessibility to soil organisms. The SOM is a dynamic and complex succession of active and stable organic fractions. The role of these fractions on aggregate dynamics and SOC storage, particularly in microaggregation, is complex. This complexity is linked to the architectural organization of mineral particles through a series of transient and persistent mechanisms of aggregation (Tisdall, 1996).

B. Organic Binding Agents of Aggregation

It is important to understand the types of organic binding agents that intervene in soil aggregation. The nature, size, strength, and configuration of aggregates depend on the action of stabilizing agents. Organic residues have their own hierarchal system in enmeshing particles and forming aggregates (Tisdall and Oades, 1982; Miller and Jastrow, 1990). Microbial polysaccharides stabilize macroaggregates, whereas humic compounds stabilize microaggregates. Initially, there is a rapid release of organic mucilages from the roots and plant debris to form macroaggregates followed by a long-term process of stabilization, which is conveyed by complex organo-mineral compounds (Greenland, 1965).

The binding agents responsible for stabilizing and arranging the aggregates are classified as temporary, transient, and persistent agents (Tisdall and Oades, 1982). Temporary agents comprise plant roots, fungal hyphae, mycorrhizal hyphae, bacterial cells, and algae. They develop simultaneously with the growth of plant roots and build up a visible organic skeleton to enmesh the mineral particles by adsorption to form young macroaggregates. Because temporary agents comprise large substances, they are mainly associated with macroaggregates, and they are greatly affected by tillage operations. Tillage diminishes the quality and quantity of the roots and microbial population, particularly in the upper horizons. Upon partial decomposition, temporary agents release fine roots, root hairs, mucilages, and other simple organic substances. Clay particles are adsorbed by these C-rich substances and placed around the decomposing materials because clay particles are more mobile than organic substances. The young residues play a major role in forming and stabilizing the macroaggregates (Skjemstad *et al.*, 1990; Cambardella and Elliot, 1993).

Transient agents consist mainly of polysaccharides and organic mucilages. Polysaccharides are non-humic compounds of high molecular weight and are easily released from organic residues and rapidly decomposed by microbial processes. They are derived from plant and animal tissues and exudations of plant roots, fungal hyphae, and bacteria (Tisdall and Oades, 1982). They represent only about 25% of humus but are the most important transient agent to aggregation (Chenu and Guerif, 1991). Polysaccharides commonly bind 10- and 50-μm aggregates through polymer bridges, and they are more readily adsorbed by clay particles than humic or fulvic acids (Martin, 1971). They are negatively charged and are relatively immobile as they interact with clay particles (Tisdall, 1996). Because of their reduced immobility, polysaccharides remain temporarily undecomposed while binding clay particles into macroaggregates. Polysaccharides also increase the modulus of rupture and the tensile strength of clay mineral agglomerates, which suggests that polysaccharides strengthen inter-particle bonds within aggregates (Chenu and Guerif, 1991). However, the effect of polysaccharides on aggregate stabilization lasts usually a few weeks because polysaccharides are easily decomposed (Wild, 1988).

Persistent agents include highly decomposed organic materials such as humic compounds, polymers, and polyvalent cations but their exact chemical composition is little understood. They are associated with microaggregation and long-term SOC sequestration. Edwards and Bremner (1967) showed that persistent compounds are in the inner portion of microaggregates adsorbed clay particles forming organo-mineral complexes through bridges of polyvalent cations. These humic compounds of high molecular weight are relatively recalcitrant and form bonds of clay-humic complexes through chelation of carboxylic or hydroxide groups with polyvalent metals (Ca^{+2}, Al^{+3}, and Fe^{+3}). Persistent agents have a long-lasting effect on microaggregate dynamics, but their role on the long C residence time in microaggregates is yet to be unraveled (Gale *et al.*, 2000a,b).

C. Influence of Land-Use on Aggregate Structure and Organic Carbon Relationships

A judicious management of croplands, forests, grasslands, and restored lands is the key to any potential C sequestration in the soil (Lal, 2002). Land-use controls the budget and transfers of C in terrestrial systems (Lal *et al.*, 2003; Smith, 2004). The magnitude of soil disturbance and the amount of residue incorporated into the soil impact aggregate and SOC dynamics. Total land area in the continental USA and Alaska of 916 million hectares (Mha) comprises 157 Mha (17.1%) cropland, 336 Mha (36.7%) grazing land, 236 Mha (25.8%) forest, 14 Mha (1.5%) Conservation Reserve Programs (CRP), and 20 Mha (2.2%) urban use (Lal *et al.*, 2003). It is estimated that the adoption of recommended management practices (RMPs) on these soils can sequester an average of 288 Tg C yr^{-1} (teragram $= 1 \times 10^{12}$ g) (Lal *et al.*, 2003). Further, Lal (2004) estimated that the global potential of SOC sequestration ranges from 0.4 to 0.8 Pg (1 Pg = petagram $= 1 \times 10^{15}$ g) C yr^{-1} in croplands, 0.2 to 0.4 Pg C yr^{-1} in forest and degraded/desertified lands, 0.01 to 0.3 Pg C yr^{-1} in rangelands and grasslands, and 0.01to 0.03 Pg C yr^{-1} in irrigated soils. The world soils can sequester between 0.4 and 1.2 Pg C yr^{-1}.

1. Forest

Forests occupy one third of the world's land area and govern C transfers through photosynthesis and respiration, representing an important C pool (Brown, 2002). Forests favor the SOC sequestration because of their increased woody biomass, extensive roots, and abundant litter (Sharrow and Ismail, 2004). The characterization of C in the aboveground biomass of forests is well advanced, but the belowground biomass dynamics in soil aggregation are poorly understood. Brown (2002) observed the need for correlating the belowground biomass to the aboveground biomass to predict C storage in forest soils. Extensive roots of forest plants influence the microbial biomass in the soil by controlling the C cycle between the atmosphere and the soil (Brown, 2002). Forests improve soil aggregation while transferring organic C into deeper soil horizons. Kaiser *et al.* (2002) determined that forest subsoil has about 45% of total SOC of the soil profile, and this fraction is bound to the clay particles forming <20 μm microaggregates. The confinement of SOC in the forest subsoils is essential for long-term storage of C because it is stabilized in the soil matrix against biological decomposition. The extent of this stabilization is determined by organo-mineral interactions, micropores, type and nature of clay surfaces, and C location within the microaggregates.

The SOC sequestration depends on forest species and management (Del Galdo *et al.*, 2003; Lal, 2003b; Lamlom and Savidge, 2003). There is a trade-off between ecosystem C storage capacity and timber production. By selecting the appropriate tree species, it is possible to increase soil's potential for C storage. Seely *et al.* (2002) estimated the long-term differences in SOC storage among three tree species including white spruce (*Picea glauca*), trembling aspen (*Populus tremuloides*), and lodgepole pine (*Pinus contorta var. latifolia*) and found that all species are important C pools. They found total C storage to be the highest in aspen, followed by that in pine and spruce. Vesterdal *et al.* (2002) showed that soils growing Norway spruce (*Picea abies* L.) sequester 9 Mg C ha^{-1} whereas those growing oak (*Quercus robur* L.) sequester only 2 Mg C ha^{-1} after 29 years and the SOC is mostly concentrated in the upper soil horizons. In Hawai, Kaye *et al.* (2000) reported that 17-year-old eucalyptus (*Eucalyptus sp.*) and siris (*Albizia lebbeck*) trees enhance SOC and N pools. Siris mixed with other species has more SOC and N concentrations than pure stands of eucalyptus or siris. Stands with the highest percentage of siris has 230 g m^{-2} more soil N and 2000 g m^{-2} more soil C than stands without siris, confirming that SOC storage depends on the tree species.

Impact of forest species on C fluxes has been widely studied. Yet, studies detailing the mechanisms of SOC storage in soil aggregates under different tree species are scanty. Del Galdo *et al.* (2003) showed that intensive agriculture significantly decreased SOC compared to permanent grassland after 20 years of management. They also showed that afforestation increased the SOC by 23% in the surface soil and improved sequestration of new and stabilization of old C within microaggregates (53–250 μm) and silt and clay (<53 μm) fractions. Jacinthe *et al.* (2001) observed the highest SOC in depositional soils followed by forest and cropland, and macroaggregates contained higher SOC concentration than microaggregates.

Reducing deforestation can decrease C emissions, especially from tropical ecosystems. Veldkamp (1994) estimated that about 2 Pg C yr^{-1} is emitted as a result of deforestation, decreasing macroaggregation, and macroporosity of the soil while reducing the soil's potential for SOC sequestration. Conversion of forests to croplands also decreases SOC concentrations and degrades soil structural properties (Lal *et al.*, 1997). Conversely, afforestation of degraded soils has a high potential of SOC sequestration depending on the tree species. Further understanding of long-term SOC storage in belowground biomass of trees in interaction with soil structure is important to ascertaining the forest soils' potential to sequester SOC.

2. *Grazing Land*

Grazing lands cover about one fifth of the world's land area (Fisher *et al.*, 1998). Soils under pasture are better aggregated and sequester more C than intensively tilled croplands because of increased return of residues and reduced soil disturbance (Percival *et al.*, 2000). Increased root biomass in pastures contributes to the storage and stabilization of SOC in aggregates. Cambardella and Elliot (1993) reported that total SOC in soil aggregates under blue grama grass (*Boutelloua gracilis* W.) was 21% in >2000 μm, 23% in 250–2000 μm, 20% in 53–250 μm, and 19% in <53 μm aggregate fractions, whereas the average SOC in bare fallow soils was only $\sim$17%. Lal (2002) observed that SOC sequestered in U.S. pastures was between 18 and 90 Tg C yr^{-1}. The high SOC concentration in pastures is attributed to the chemical stabilization of organic C in the soil matrix (Percival *et al.*, 2000).

Soil aggregation dynamics may differ between grasslands and forests. Sharrow and Ismail (2004) estimated that about 90% of organic C in pastures is stored belowground, whereas $>$60% of organic C in forests is stored aboveground. The high belowground residue in pastures may enhance aggregation. The organic C stored in grasslands is in greater interaction with the soil than in forests because most of the SOM in grasses is belowground. The increased soil and SOM interaction in grasslands can promote long-term SOC sequestration within the microaggregates compared to forests, but differences in total C storage between grasslands and forests are often mixed. The C storage in grasslands can be equal (Corre *et al.*, 1999), higher (Garten and Ashwood, 2002), or lower (Silver *et al.*, 2000; de Koning *et al.*, 2003) compared to that in forests.

The effect of pastures on SOC sequestration differs among species. Six *et al.* (2001) found higher SOC concentrations in macroaggregates under white clover (*Trifolium repens*) than under perennial ryegrass (*Lolium perenne*), indicating a rapid macroaggregate turnover in white clover due to higher residue quality and weaker root mass response. Min *et al.* (2003) reported that soils under alfalfa (*Medicago sativa* L.) sequester more SOC than those under orchardgrass (*Dactylis glomerata* L.) because of high soil aggregation under alfalfa. Differences in root mass (Hebeisen *et al.*, 1997) and litter composition (Fisher *et al.*, 1998) explain differences in SOC sequestration among grass species. The SOC sequestration in grazing lands can be increased by improving forage quality and productivity (Fisher *et al.*, 1998) and by rehabilitating overgrazed grasslands (Conant and Paustian, 2002). There is a need, however, of more research to elucidate the mechanisms responsible for SOC storage in aggregates in soils under pasture (Six *et al.*, 2000).

3. *Cropland*

Croplands may serve either as a sink or source for atmospheric CO_2 (Lal *et al.*, 1998). The SOC sequestration in croplands is controlled by balance between the C in residues returned to soil and that released into the atmosphere. Cropping and tillage systems that promote the return of residues to the soil increase the SOC pool and aggregation (Gale and Cambardella, 2000). Agricultural lands in the United States can potentially sequester 30–105 Tg C yr^{-1} (Follet, 2001).

3.1 Tillage Practices. Tillage practices impact the mechanisms of SOC sequestration in aggregates, but their influence depends on the magnitude of soil disturbance (Figure 1). For example, no-till (NT) conserves organic matter and stabilizes aggregates while increasing SOC pool (Lal *et al.*, 1998; Lal, 2002). No-till and reduced till promote C-enriched macroaggregation (Skidmore *et al.*, 1986; Gale and Cambardella, 2000; Six *et al.*, 2000). Because of low macroaggregate turnover, NT soils sequester more C than conventional till (CT) soils (Six *et al.*, 1999a; Table 1; Figure 1). McKonkey *et al.* (2003) found that NT sequesters 67–512 kg C ha^{-1} yr^{-1} more than CT. Increased plant residues left in NT as compared to CT increase the biological activity, resulting in greater macro- and microaggregation to physically shelter the SOC within the aggregates. No-till may have twice more stable and free microaggregates than CT soils, promoting SOC encapsulation within the microaggregates due to slower macroaggregate turnover (Six *et al.*, 2000). The SOC-rich residues in microaggregates are protected from microbial and enzymatic actions, thereby remaining relatively undecomposed for a long time. A factor for soil aggregation in NT is the abundant microbial biomass including bacteria, fungi, and especially the mycorrhizal fungi compared to CT (Frey *et al.*, 1999). Thus, NT practices are being recommended as a strategy to reduce C depletion from agricultural lands. However, effects

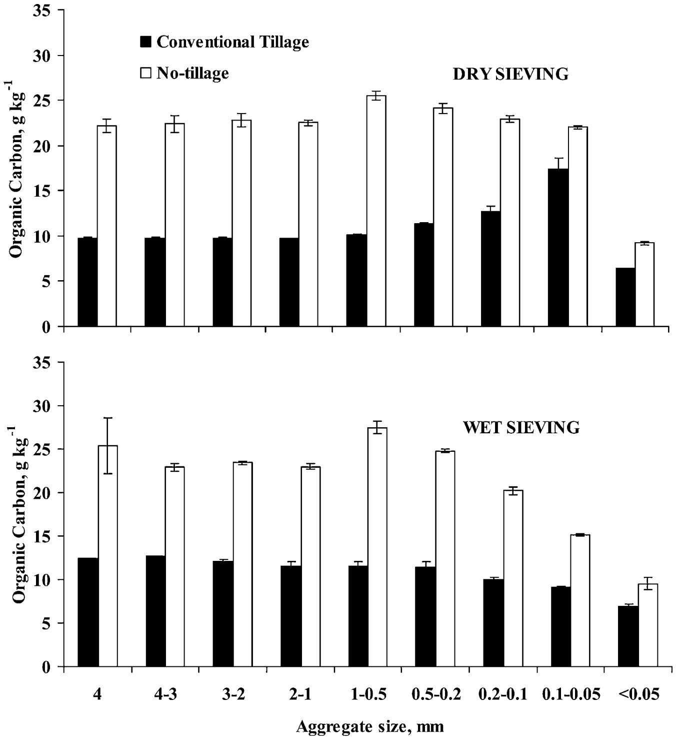

FIG. 1. Relationship of soil organic carbon (SOC) and macro- and microaggregates as affected by sieving techniques and by management (Redrawn from Puget *et al.*, 1995).

of NT management on aggregate dynamics in interaction with the type of organic matter have been little studied (Six *et al.*, 2000).

The CT practices accelerate the aggregate turnover (Bossuyt *et al.*, 2002) and increase the decomposition of organic residues (Six *et al.*, 2000), thereby reducing SOC concentration (Lal, 2002). Tillage disrupts aggregation, exposes existing aggregates to microbial processes, accelerates organic C turnover, and reduces SOC accumulation (Table 1). Tillage changes the natural ^{13}C content and concentration of SOC. Table 2 presents the effect of tillage on C enrichment based on natural ^{13}C abundance for different aggregate sizes. The more negative the ^{13}C values, the lower the enrichment in ^{13}C (Table 2). No-till soils can be equal (Six *et al.*, 2000) or richer (Puget *et al.*, 1995) in ^{13}C compared to CT. According to Puget *et al.* (1995), macroaggregates tend to be more enriched in ^{13}C than microaggregates in both NT and CT soils (Table 2).

Intensive tillage leads to fewer stable microaggregates (Puget *et al.*, 1995). Further, drastically disturbed soils are prone to physical degradation, which reduces the re-aggregation of soil around the SOC (Denef *et al.*, 2001). The processes of SOC and soil structure interaction cease when the system is disrupted by cultivation, leading to the formation of C-depleted microaggregates. Tilled soils not only create a less conducive environment for natural aggregation but also cause frequent instability of macroaggregates (Paustian *et al.*, 1999). Further, tillage alters the SOC distribution within both the soil profile and aggregates. In a 20-year rotation in southern Canada, Yang and Kay (2001) showed that SOC in the upper 0–10 cm layers was higher than in the 10–20 cm soil depth in chisel plow but the opposite occurred in moldboard plow soils. Moldboard plow often incorporates residues to deeper layers and reduces macroaggregation.

Tillage effects on the degradation of macro- and microaggregates are rather selective. Microaggregates often remain relatively intact during tillage compared to macroaggregates (Beare *et al.*, 1994a; Denef *et al.*, 2001) because they are bound by resistant binding agents (Tisdall and Oades, 1982; Six *et al.*, 1999b; Bossuyt *et al.*, 2002). Research shows that SOC sequestration in the soils is a function of soil structural stability (Angers and Chenu, 1998; Six *et al.*, 1999a). Therefore, any cultivation practice that reduces the disruption of aggregates will enhance SOC sequestration.

3.2 Rotations and Cropping Systems. Crop rotations influence soil aggregation and thus SOC sequestration. Crop rotation effects on SOC sequestration are, however, altered by tillage (Lal, 2002), quantity and quality of organic residues (Chan and Heenan, 1999), type of crops (West and Post, 2002), and site-specific soil characteristics (Lal *et al.*, 1997; Jarecki and Lal, 2003). Crop rotations integrated with NT are conservation-effective practices that promote SOC storage (Aoyama *et al.*, 1999; Lal, 2002) and macro- and microaggregation to sequester C (Hernanz *et al.*, 2002). Macroaggregation and aggregate stability are often higher in NT than CT-based rotation systems (Filho *et al.*, 2002). Whalen *et al.* (2003) reported that 2-year NT soils in corn-soybean (*Glycine max* L.) rotation sequester more SOC and have higher water stable aggregates (>4 mm) than any rotation of crops in CT soils.

The effectiveness of crop rotations in sequestering SOC depends on the cropping system and tillage practices. West and Post (2002) using a global data of SOC concluded that crop rotations sequester an average of 20 g C m^{-2} yr^{-1} if continuous corn (*Zea mays* L.) and corn–soybean cropping systems are excluded. In Canada, Yang and Kay (2001) found that 2-year rotation of barley (*Hordeum vulgare*) followed by 2-year corn sequestered the least amount of SOC, whereas soils under soybean plus winter wheat (*Triticum aestivum* L.) followed by 2-year corn had more SOC sequestered than continuous corn following 2-year of alfalfa or soybean. Intensifying crop rotation practices and eliminating summer fallow increases SOC concentration (West and Post, 2002; Jarecki and Lal, 2003). McKonkey *et al.* (2003) reported that diverse crop rotations sequester 27–430 kg C ha^{-1} annually, and this amount is more than rotations that include bare fallow.

The inclusion of legumes in crop rotations changes the dynamics of macro- and microaggregation for SOC storage. Residues of legumes are often rich in labile organic fraction, which increases soil aggregation and SOC concentration but this effect is often transient because the labile fraction is easily degraded. Wright and Hons (2004) indicated that soils under grain sorghum (*Sorghum bicolor* L.)/wheat/soybean rotations sequester more SOC and organic N than those under continuous soybean. Labile organic fractions are often found in transient

TABLE 1
Relationship of soil organic carbon with macro- and microaggregates as affected by soil and crop management

Location	Soil	Tillage[1]	Aggregate size distribution (mm) Organic C, g kg^{-1}							Reference
			>3	3–2	2–1	1–0.5	0.5–0.2	0.1–0.05	<0.05	
Sidney, NE	Loam	NT	—	28	—	—	34	31	19.5	Cambardella and Elliot (1993)
		Fallow	—	25	—	—	24	22.5	18.5	
Athens, GA	Sandy clay loam	NT	—	81	—	106	131	87	25	Beare *et al.* (1994a)
		CT	—	37	—	50	54	36	23	
Batavia, IL	Silt loam	Corn	2	2	1	3	5	12	3	Jastrow *et al.* (1996)
		Pasture	5	15	10	5	3	2	2	
Quebec, Canada	Silty loam	Corn	—	25	28	29	24	22	30	Angers and Giroux (1996)
		Pasture	—	26	29	30	32	27	27	
		Rotation	—	28	30	30	26	24	35	
Boigneville, France	Silty clay	NT	22	23	23	26	24	22	9	Puget *et al.* (1995)
		CT	10	10	10	10	11	17	6	
Canada	Loam	NT	18	—	—	24	13	—	15	Franzluebbers and Arshad (1997)
		CT	20	—	—	29	14	—-	18	
	Silt loam	NT	25	—	—	29	18	—	10	
		CT	26	—	—	33	20	—	12	
	Clay loam	NT	32	—	—	34	34	—	18	
		CT	30	—	—	32	32	—	20	
	Clay	NT	35	—	—	35	35	—	30	
		CT	33	—	—	32	34	—	30	
Columbia, MO	Silt loam	Soybean	—	—	22	18	17	22	12	Buyanovsky *et al.* (1994)
Brazil	Clay	Forest	—	—	26	24	27	31	33	Lehmann *et al.* (2001)

[1] NT, no-till; CT, conventional tillage.

TABLE 2
Carbon sequestration in different aggregate sizes based on isotope ratios (δ^{13}C)

Location	Soil	Tillage[1]	Aggregate size distribution (mm) ^{13}C							Reference
			>3	3–2	2–1	1–0.5	0.5–0.2	0.1–0.05	<0.05	
Sidney, NE	Loam	NT	—	—	−21.7	−21.4	−23.6	−20.6	−19.9	Six *et al.* (2000)
		CT	—	—	−22.0	−20.3	−22.7	−19.2	−18.5	
France	Loam	Corn	—	—	—	—	−24.3	—	−26.3	Besnard *et al.* (1996)
		Forest	—	—	—	—	−27.6	—	−27.5	
Batavia, IL	Silt loam	Corn	−17.2	−17.3	−17.1	−17.2	−17.4	−17.8		Jastrow *et al.* (1996)
		Pasture	−21.4	−21.7	−21.2	−20.4	−20.8	−21.6		
Quebec, Canada	Silty loam	Corn	—	−26.0	−25.6	−26.0	−25.7	−26.1	−26.2	Angers and Giroux (1996)
		Pasture	—	−27.1	−27.1	−27.4	−27.3	−27.2	−27.3	
		Rotation	—	−26.9	−25.6	−26.0	−25.7	−26.1	−26.2	
Ontario, Canada	Silty clay to clay loam	Corn	—	—	—	—	−27.5	−23.7	−24.7	Monreal *et al.* (1997)
		Forest	—	—	—	—	−23.5	−27.4	−27.2	
Boigneville, France	Silty clay	NT	−16.5	−16.6	−16.7	−16.4	−16.7	−17.4	−19.4	Puget *et al.* (1995)
		CT	−20.1	−19.9	−20.2	−20.1	−20.2	−20.7	−21.3	

[1] NT, no-till; CT, conventional tillage.

macroaggregates and their effect on long-term SOC sequestration is questionable. Studies relating differences in organic matter quality to formation and stabilization of macro- and microaggregates under various cropping rotations are few (Chan and Heenan, 1999). Fractions of organic matter that correlate the most with changes in macro- and microaggregate turnover are yet to be identified under different crops in rotation.

3.3 Manuring. Manuring improves soil quality and SOC pools (Whalen and Chang, 2002). Formation of aggregates and SOC concentration is, for example, enhanced under dairy manuring (Min *et al*., 2003). Manuring increases both the POM and mineral associated organic C in macroaggregates, increasing the amount of C sequestered (Jenkinson and Rayner, 1977; Figure 2). Increased POM concentration from manuring stabilizes macroaggregation and increases their resistance to slaking. Manure-derived organic materials are greater in NT than in tilled systems due to differences in mineralization rates (Aoyama *et al*., 1999). In some cases, manuring can even increase macroaggregate-protected SOC in tilled systems (Schjonning *et al*., 2002). Organic matter in manure entering the soil is initially in the POM form and then converted into mineral-associated SOC pool (Aoyama *et al*., 1999). Schjonning *et al*. (2002) found that SOC concentration is higher in organically managed than in conventionally managed dairy farm soils, and it correlates linearly with macropores while promoting biotic mechanisms responsible for macroaggregation. Organic amendments are beneficial to proper disposal of dairy manures in forage systems while increasing soil potential for SOC sequestration (Min *et al*., 2003). Well-planned nutrient management and manuring are vital to SOC sequestration in agricultural lands.

3.4 Nutrient Management. Efficient nutrient management is essential to the humification of C in plant residues by soil organisms. Aggregation depends on the rate of humification as mineral particles cluster around humified substances while sequestering SOC. Enhancement of soil fertility increases microbial biomass and C pools. Inorganic fertilization (N, P, S) may be important to enhancing humification of C in residues and SOC sequestration (Lal, 2003a,b). Nitrogen fertilization alone may (Campbell *et al*., 2000) or may not (Halvorson *et al*., 2002) improve SOC sequestration. Seely *et al*. (2002) reported that N fertilization increases both the productivity of aspen and SOC sequestration. It increases quantity of crop residues in the Great Plains, but its effect on SOC sequestration is questionable (Halvorson *et al*., 2002). Min *et al*. (2003) studied the effect of 5-year fertilization in alfalfa and orchardgrass forage systems and found that continuous use of inorganic fertilizers does not improve soil aggregation nor enhance SOC sequestration compared to dairy manuring.

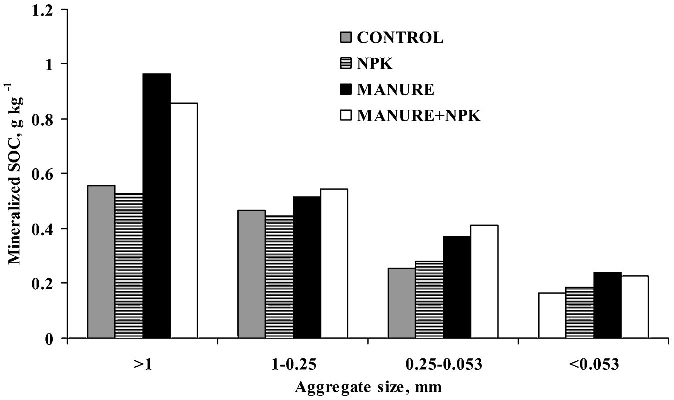

FIG. 2. Mineralized soil organic carbon (SOC) content under different sized aggregates in response to manuring and inorganic fertilization (N, nitrogen; P, phosphorus; K, potassium; Redrawn from Aoyama *et al*., 1999).

There is an increasing trend of using organic rather than inorganic fertilizers due to concern of food safety, environmental pollution, and the urgency to dispose of animal and municipal wastes. Combined use of organic fertilizers with green manures and crop rotations can be a suitable nutrient management alternative (Figure 2). Nutrient management involves an understanding of sources of organic nutrients in relation to C and N. Soils under corn-corn-soybean-wheat rotation, continuous corn, and cover crops receiving compost had 14% higher SOC than those receiving fertilizers but there occurred a deficit of N for optimum C storage (Fortuna *et al*., 2003). Dersch and Bohm (2001) determined that a 36-year N fertilization increased SOC pools by an average of 2.1 Mg ha^{-1} compared with no fertilization. The SOC pool increased by about 5.6 Mg ha^{-1} after 21 years when farmyard manure was applied at 10 Mg ha^{-1} yr^{-1}. These studies suggest that management of nutrients by using inorganic fertilizers, manure, compost, cover crops, and crop rotations is essential not only to maintaining soil fertility but also to increasing SOC pools.

3.5 Irrigation. Irrigation is important to enhancing agricultural production in arid and semi-arid regions. Increased SOM content in irrigated soils can increase aggregation. Under long-term management, there are more water stable aggregates in irrigated than in dryland soils. Further, irrigated soils have a relatively high SOC pool (Whalen and Chang, 2002). Supplemental irrigation has the potential to enhance SOC sequestration by 50 to 150 kg ha^{-1} yr^{-1} in agricultural soils (Follett, 2001). Irrigation in non-agricultural ecosystems can also be beneficial to SOC sequestration. Mancino and Pepper (1992) observed that irrigation of turf soils under bermudagrass (*Cynodon dactylon* L.) significantly increased SOC concentration. Similarly, C accumulation in trees increased by 25% due to irrigation and by 37% due to fertilization with ammonium sulphate (100 kg N, 114 kg S ha^{-1} yr^{-1}), whereas simulated drought decreased SOC stock by 15% (Nilsson, 1993). Irrigation combined with improved inorganic fertilization (Nilsson, 1993) and manuring (Whalen and Chang, 2002) can promote macroaggregate formation and stabilization while significantly sequestering SOC and reducing the net CO_2 emissions.

4. *Urban Land*

Urban encroachment is a continuing phenomenon in accord with the population growth. City dwellers are demanding more and more land for urban purposes in the United States and worldwide. Turfgrass in home lawns, schools, parks, recreational facilities and commercial and industrial sites occupies about 20 million ha of land in the United States, which is about 27% more than the land (14.7 ha) enrolled under USDA Conservation Reserve Program (CRP) (Qian and Follet, 2002). This land under lawns impacts the C cycle as a sink of atmospheric CO_2. Pouyat *et al.* (2002) found that urban forests have higher SOC than suburban and rural areas. They also showed that SOC in institutional land and low-density residential areas is 38 and 44% higher than that in commercial lands, respectively. Intensive urban land use diminishes the soil's ability to sequester SOC. Nowak and Crane (2002) used field data from 10 U.S. cities and national urban tree cover data and estimated that urban trees in the United States store 700 Tg of C with a total SOC sequestration rate of 22.8 Tg C yr^{-1}. Carbon storage within cities ranges from 1.2 Tg C yr^{-1} in New York, NY to 19,300 Tg C yr^{-1} in Jersey City, NJ. Regions with the greatest proportion of urban land are the Northeast (8.5%) and the southeast (7.1%). Urban forests in the north central, northeast, south central, and southeast regions of the United States sequester more SOC than other regions. Conversion of non-urban to urban lands greatly modifies SOC pools and dynamics, but data on urban ecosystems to assess SOC fluxes in interaction with soil aggregates are limited (Qian *et al.*, 2003).

5. *Recreational Land*

Recreational lands are also continually expanding along with urbanization. These lands include golf courses, parks, sport fields, and so on and are an important component of the terrestrial ecosystem. However, the role of recreational lands on SOC sequestration has not been studied extensively. Qian and Follett (2002) analyzed 45-year data collected from turfgrass management in golf courses and reported that SOM responds linearly to turfgrass culture. The most rapid increase in SOC sequestration occurs in the first 25 to 30 years of turfgrass establishment storing about 1.0 Mg C ha^{-1} yr^{-1} for both fairways and putting greens. The amount of SOC sequestration in golf courses can be comparable to that sequestered in lands under CRP. The increased contribution of turfgrass to sequester atmospheric CO_2 is attributed to the high amount of root biomass and lack of soil disturbance of turf soils. Turfgrasses improve soil aggregation through the addition of SOM and deep rooting system (0.5 to 3.0 m). Warm season grasses are more beneficial than cool season grasses in sequestering SOC and improving soil quality in recreational areas (Beard and Green, 1994) because of deeper and extensive roots. There is a need to assess the rate and ability of turfgrass soils for long-term storage of SOC and soil structure improvement under different turfgrass management systems.

D. Soil Biota and Soil Carbon Dynamics

Soil structure is a complex mesh of pathways and galleries inhabited by millions of microorganisms each conducting a specific function relative to soil aggregation and SOC distribution and protection. There is a two-way interaction of aggregate structure and biota relations. Soil biota are crucial to formation of aggregates and, in turn, aggregates provide habitat for soil biota. Microaggregates develop in zones of high microbial activity and high production of humic substances (Oades and Waters, 1991). Plant roots, both alive and dead, can congregate a large population of colonizing microorganisms. Soil biota are responsible for the stabilization and turnover of SOC in aggregates. Interactions of SOC with aggregates are mediated by soil biota including fungi, bacteria, plant roots, earthworms, and other soil organisms (Oades, 1984). These organisms control the decomposition of organic residues, transformation of C, and SOC sequestration in aggregates (Franzluebbers and Arshad, 1997).

The scale at which soil organisms act depends on the species. Earthworms and termites are associated with the formation and stabilization of macroaggregates, whereas fungi and bacteria commonly form microaggregates. Earthworms consume soil and SOM and form macroaggregates, which are richer in SOC and microbial population than the original material (Tisdall, 1996). Earthworms impact microaggregation by mixing the soil with SOM inside their digestive tracts and promoting the interaction of soil particles with microflora. Jégou *et al.* (2000) found that *Lumbricus terrestris* and *Aporrecodea giardi* increase aggregation and enhance SOC accumulation in casts.

In contrast, fungal hyphae can extend over soil particles and aggregates, forming a mat type mesh or network of hyphae inside the aggregates. Hyphal length can grow up to 10 mm on the surface of roots to stabilize macroaggregates. Tisdall *et al.* (1997) observed that fungal hyphae including saprophytic and mycorrhizal fungi bind individual soil particles and organic materials to form <2 μm microaggregates and then enmesh these microaggregates into >50 μm aggregates by secreting polysaccharides and other by-products. Jastrow *et al.* (1998) found a high correlation ($r = 0.9$) between macroaggregate and hyphal length. Bacteria often accumulate within soil aggregates and are transported rapidly by water flow (Oades, 1984). Because of abundant micropores, clay soils are usually rich in bacteria and fungi, whereas sandy soils often have low microbial population due to reduced SOM and increased macroporosity. Biological diversity of soil fauna defines the sequential steps of aggregation in direct correlation with soil type and organic matter.

II. COMPOSITION OF PLANT RESIDUES AND AGGREGATES

A. C:N Ratio

The ratio of carbon:nitrogen (C:N) of plant residues determines the effectiveness of organic material for improving soil structure and thus enhancing C sequestration. Organic residues with higher C:N ratio persist for long times and improve soil aggregation (Hagerdon *et al.*, 2003). Residues with low C:N ratio are rapidly decomposed and have reduced impact on soil structure improvement. For example, soybean residues have lower C:N ratio and are more rapidly decomposed than wheat residues (Ghidey and Alberts, 1993). The C:N ratio is variable in organic residues. It is 13 in freshly cut alfalfa, 80 in wheat straw, and 90 in fresh tree leaves (Buyanovsky *et al*., 1994; Tisdall, 1996). Hu *et al*. (2001) determined that increased atmospheric CO_2 increases microbial biomass in the soil, thus raising the N uptake by plants. This reduces N concentration for microbial use, decreasing the microbial activity and SOM decomposition and thus increasing the SOC sequestration. High C:N ratio commonly increases the potential of the soil as a sink for atmospheric CO_2. The dynamics of C:N are complex and vary between soils and within the same soil. Saroa and Lal (2003) found that C:N ratio decreased with decreasing particle size and with increasing soil depth.

Soils under NT usually have higher C and N concentrations than soils under CT. Wright and Hons (2004) reported that NT improved sequestration of both SOC and organic N in macroaggregates compared to CT because NT had significantly lower C:N ratio. Rapidly decomposing residues enhance the formation of aggregates, but their action is transient, whereas slowly decomposing residues have a gradual impact on aggregation and their long-term effect on SOC sequestration is higher than rapidly decomposing residues. Because of the importance of C:N ratio to SOC sequestration in soils, the dynamics of N in macro- and microaggregates need to be taken into account for addressing the potential of soils in SOC sequestration.

B. Lignin Content and Phenolic Compounds

The effect of organic residues on soil aggregation is a function of the biochemical properties of the residues including lignin, phenolic compounds, aromatic compounds, sterols, and lipids concentrations. Lignin content and C:N ratio are residue parameters that affect the decomposition of SOM. The SOC concentration is positively correlated with lignin content (Lamlom and Savidge, 2003). Early or physical decomposition of residues depends on the C:N ratio (Melillo *et al*., 1982), but advanced or chemical decomposition of organic residues is controlled by lignin concentration (Martens, 2000). Mature organic materials with high lignin content are important to soil structure development. Lignin is mostly associated with the macroaggregation (Monreal *et al*., 1997) and stabilizes and binds particles into aggregates (Magill and Aber, 1998). Lignin at the surface of the aggregates is more decomposed than that within the aggregates due to increased microbial activity outside, which explains the greater SOC sequestration within microaggregates (Amelung *et al*., 2002). Long residence times of lignin-rich materials can enhance long-term SOC sequestration in aggregates.

Phenolic compounds decompose at slower rates than polysaccharides and amino acids and can promote long-term aggregation (Martin, 1971). Indeed, Martens (2000) reported that aggregate stabilization is highly correlated ($r = 0.96$) with phenolic acid concentration in native prairie and in corn–soybean rotation systems. Tillage lowers the concentration of phenolic compounds, resulting in decreased aggregate stability. Aggregates stabilized by temporary binding agents are further strengthened by phenolic compounds. Stable humic compounds (*i.e*., phenolic compounds) are far more important to long-term regulation of SOC sequestration in aggregates than unstable organic residues.

III. MODELS OF SOIL AGGREGATION AND ORGANIC CARBON DYNAMICS

Several theories and conceptual models have been proposed to relate soil aggregation to SOC dynamics (Emerson, 1959; Greenland, 1965; Tisdall and Oades, 1982; Gale *et al*., 2000b; Six *et al*., 2000; Table 3). Most of these models proclaim that soils consist of ever-changing aggregates of different sizes bound together by organic and inorganic compounds. Development of the conceptual models of aggregate organization has enhanced our understanding of aggregate dynamics for sequestering SOC, but a full understanding of the mechanisms of aggregate dynamics in relation to SOC sequestration is awaiting further development (Gale *et al*., 2000a; Six *et al*., 2000; Bossuyt *et al*., 2002). Some discrepancies exist between classical and modern concepts with respect to the formation of macro- and microaggregates. Furthermore, classical models were mainly developed to explain the processes of soil aggregation that are important to plant growth and biological activity. Current conceptual models of soil aggregation are being directed to elucidate concerns about SOC sequestration in aggregates.

A. Classical Models

Emerson (1959) postulated a simplified model, which indicates that organic substances bridge sand and clay crystals to form and stabilize aggregates or what he called "soil crumbs." The model states that the number and nature of the bonds determine the aggregate stability. According to this model, the amount of organic C stored in aggregates is directly proportional to the specific surface area of clay minerals. Greenland (1965) stated that aggregates are formed and stabilized by microbial polysaccharides derived from fresh plant residues, but the action of polysaccharides depends on soil intrinsic characteristics. Edwards and Bremner (1967) argued that formation of macroaggregates results from a systematic linkage of microaggregates and these are the product of bonding of $<2\ \mu$m microstructures consisting of organo-mineral complexes. This model thus suggests that bonds within the microaggregates are much stronger than those between the microaggregates forming blocks of macroaggregates.

Tisdall and Oades (1982) proposed a pioneering conceptual model for aggregate hierarchy in which SOM is the principal binding agent of aggregate formation (Figure 3). The concept behind this system is that binding of microaggregates is responsible for the formation of macroaggregates. Primary particles and then microaggregates are linked together to form macroaggregates. The model proposes four stages of

TABLE 3
Mechanism models for soil organic carbon (SOC) sequestration in soil aggregates in a chronological sequence

Location	Soil	Mechanisms of SOC sequestration	Reference
England	Clay	Organic matter acts like a cement during soil aggregation by releasing humic and other organic compounds.	Russell (1938)
Australia	Clay	Organic matter is protected between clay and sand particles. Clay surface area and the resistance of organo-mineral attractions to microbial processes control storage of SOC.	Emerson (1959)
Ames, IA	Clay loam	Bonding of <2 μm microstructures formed through clay particle-polyvalent metal-organic matter reactions creates microaggregates to form macroaggregates.	Edwards and Bremner (1967)
Australia	Fine sandy loam	Clay minerals form microaggregates. Organic materials are the bridging agents that bind clay, silt, and sand particles with microaggregates to form macroaggregates.	Greenland (1979)
Australia	Fine sandy loam	C-enriched persistent organic agents bind free primary particles into microaggregates and then microaggregates form macroaggregates through temporary and transient agents. Upon degradation, macroaggregates free stable microaggregates.	Tisdall and Oades (1982)
Australia	Fine sandy loam and clay	Roots and fungal hyphae enmesh >250 μm macroaggregates and, by disruption, macroaggregates release 20 to 250 μm microaggregates, and before freeing <20 μm particles sequestering organic C molecules.	Oades (1984)
Athens, GA	Loam	Stable and C-enriched particulate organic matter is sequestered in the largest (53–106 μm) microaggregates formed within macroaggregates. Microaggregates promote long-term encapsulation of organic C in contrast with macroaggregates.	Beare *et al.* (1994a)
Australia	Fine sandy loam	Microbes colonize plant and root debris and form >250 μm macroaggregates, which gradually release stable 20 to 250 μm microaggregates. These microaggregates free stable <20 μm microaggregates formed by clay-humic complexes.	Golchin *et al.* (1998)
Sidney, NE	Loam	C-enriched fresh residues build macroaggregates and then become inter-aggregate particulate organic matter to build microaggregates within macroaggregates, which later liberate free microaggregates for the next cycle of macroaggregation.	Six *et al.* (2000)
Athens, GA	Loam	Total C is high in no-till macroaggregates, but long-term C protection mainly occurs within microaggregates. Tilled soils store more total and young C than no-till in the subsurface horizons but its protection is short.	Bossuyt *et al.* (2002)

macro- and microaggregation (Figure 3). The stages are not fixed, and the aggregation is far from being smooth and sequential, particularly for small aggregates. Microaggregates (<2 μm) are comprised of clay plates that are linked through ionic charges of organic particles, whereas those between 2 and 20 μm are bound solely by persistent agents. Aggregates (20–250 μm) are linked by the interaction of transient and persistent agents in association with aluminosilicates. Large and relatively unstable aggregates (>1000 μm) are enmeshed by temporary agents such as roots and fungal hyphae in soils with $>2\%$ of SOM, and by transient agents in soils with $<2\%$ of SOM. Inorganic binding agents including alumino-silicates and hydrous oxides of Al and Fe also intervene in the formation of large aggregates but their action is relatively small.

B. Contemporary Models

Most modern approaches of aggregate organization challenge the hierarchal model by Tisdall and Oades (1982), which hypothesized that microaggregates are formed first before building macroaggregates. Oades (1984) revised this model of aggregate hierarchy and indicated that macroaggregates (>250 μm) burst into microaggregates (20 to 250 μm) as a prior step in forming microaggregates (<20 μm; Figure 3). According to this model, microaggregates are primarily formed within macroaggregates.

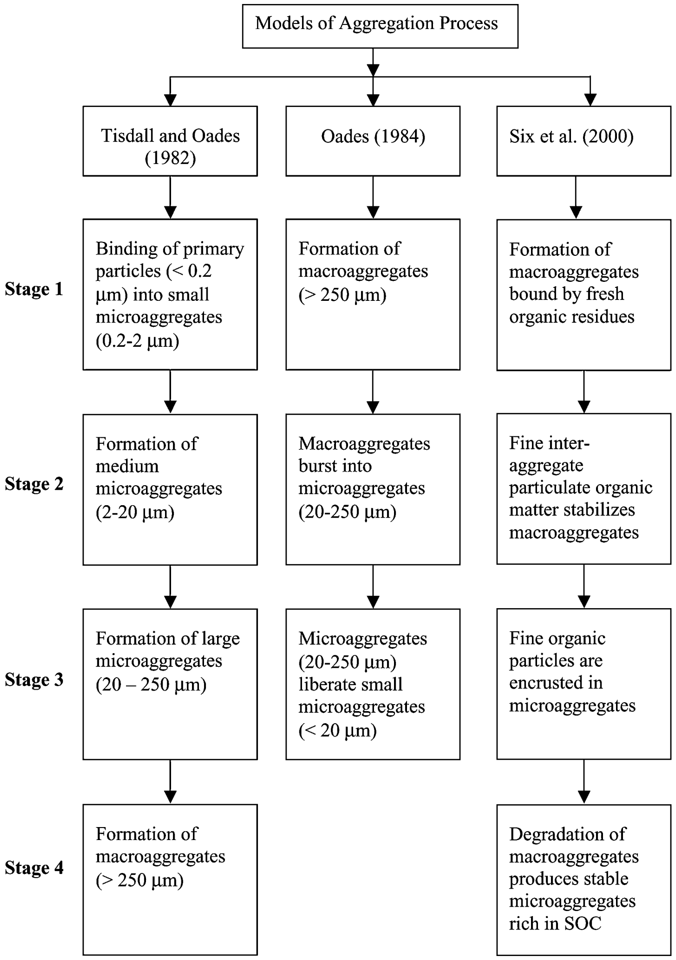

FIG. 3. Selected models of macro- and microaggregation of soils.

Most of the contemporary research in the relations of soil structure and SOC relies on the model by Oades (1984). Golchin *et al.* (1998) proposed a model similar to that of Oades (1984) where unstable and fresh macroaggregates break down in medium microaggregates (<250 μm) and then into small microaggregates (<20 μm) occluding fine POM in their interior. The model by Golchin and colleagues, however, stresses the importance of POM to aggregation. The POM is the active pool of SOM comprised mostly of coarse and fine unstable organic detritus in various stages of decomposition and constitutes between 10–20% of the total SOM. It differs from the passive organic pool because it is readily decomposable. Free POM from coarse residues in contact with the soil adsorbs soil particles through microbial colonization, forming clusters of macroaggregates. The POM fraction within microaggregates is protected from microbial decomposition (Golchin *et al.*, 1998).

Another contemporary model by Six *et al.* (1999b) identifies four dynamic stages of macroaggregate turnover, microaggregate formation, and SOC stabilization in microaggregates (Figure 3). The model indicates that macroaggregation is stabilized by fresh plant debris and roots and by fine inter-aggregate POM. The fine organic particles are sequestered in the microaggregates formed inside the macroaggregates. Stable microaggregates are released and then build new macroaggregates. Six *et al.* (1999b) also tested the previous models and found that aggregate hierarchy does exist, particularly in soils with 2:1 clay minerals. Contemporary models (Golchin *et al.*, 1998; Six *et al.*, 1999b; Jastrow, 1996; Jastrow *et al.*, 1996) corroborate the basic principles of the model proposed by Oades (1984).

Current state-of-the-knowledge based on the existing models leads to the concept that C sequestration is a function of the architectural system of aggregate packing. The SOC turnover decreases from macro- to microaggregates, thereby implying that there is a greater physical protection of SOC in microaggregates, which could be translated into greater SOC sequestration. The SOC in microaggregates is older than that in macroaggregates because of longer storage and less disruption. The SOC is first enmeshed in microaggregates and then emerges as a dynamic nucleus for the formation of macroaggregates. The SOC is essential to the formation of macroaggregates because it is a primary source of energy for the microorganisms responsible for binding soil particles (Oades and Waters, 1991; Tisdall *et al.*, 1997; Six *et al.*, 1999b). Macroaggregates promote greater storage of SOC than microaggregates (Angers and Carter, 1996; Jastrow and Miller, 1998; Puget *et al.*, 1995), but this storage is transient (Franzluebbers and Arshad, 1997; Sainju *et al.*, 2003). Microaggregates, in contrast, promote long-term SOC sequestration (Monreal and Kodama, 1997), implying that C sequestration would decrease with increasing aggregate size. There is complexity associated with the interaction of microaggregates and SOC. Duiker and Lal (1999) acknowledged the complexity of soil aggregation and suggested that a full understanding of the mechanisms of SOC dynamics in aggregates is needed. The modified model by Oades (1984) is the basis of current studies of understanding the soil structure and SOM dynamics. However, a unique model of aggregate hierarchy that could explain all the mechanisms of macro- and microaggregation and stabilization and turnover of SOC in all aggregates is awaiting further development.

Available models cannot explain the aggregate hierarchy, in all soils, and are mainly valid in soils where SOM is the major binding agent (Tisdall and Oades, 1982; Oades, 1984). Aggregation depends on soil taxonomic class in association with SOM content where Oxisols, for example, produce water-stable aggregates but fail to produce an aggregate hierarchy because the main aggregate stabilizing agents are hydrous oxides of Al and Fe (Oades and Waters, 1991). Macroaggregate breakdown in Oxisols leads to a rapid release of <20 μm particles generating 20–250 μm microaggregates. In contrast, Mollisols and Alfisols promote the hierarchal macroaggregation owing to the abundant network of coarse roots and fungal hyphae proliferation where small organic fragments are entrapped in <20 μm microaggregates reducing their microbial decomposition, and stabilized by clay microstructure and humic complexes.

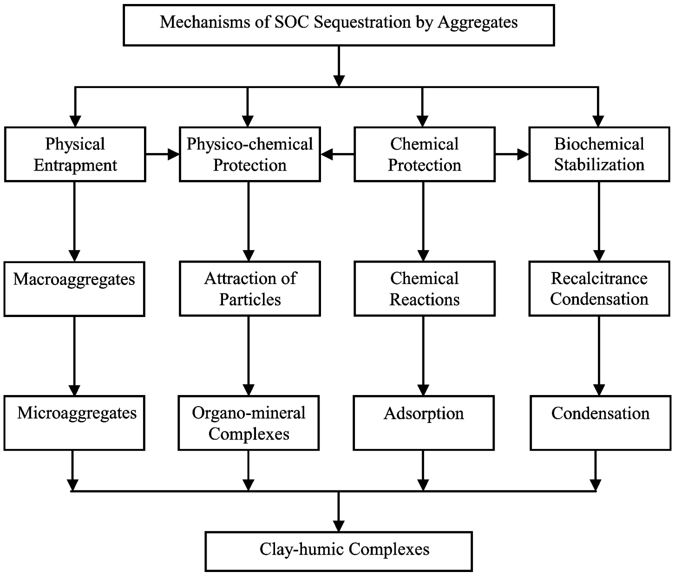

FIG. 4. Pathways of soil organic carbon (SOC) sequestration by aggregates (Emerson, 1959; Edwards and Bremner, 1967; Tisdall and Oades, 1982; Golchin *et al.*, 1994; Laird, 2001; Six *et al.*, 2002).

C. Protection and Residence Time of Soil Organic Carbon within Aggregates

The SOC sequestration in the soil is governed by the degree of physical, chemical, and physicochemical stabilization of SOM inside the aggregates (Figure 4). Physical protection refers to the entrapment of organic particles in macro- and microaggregates, whereas the chemical protection involves adsorption and recalcitrance of organic particles. Physicochemical interactive mechanisms define the maximum SOC sequestration capacity in a soil (Jastrow and Miller, 1998; Six *et al.*, 2002). The encrustation of SOM in the center of microaggregates is the fundamental pathway to SOC sequestration (Tisdall and Oades, 1982; Golchin *et al.*, 1994; Jastrow *et al.*, 1996). This process of encrustation prevents organic matter from physical and chemical decomposition by microbial processes while sequestering SOC. The protected SOC pool stabilizes the microaggregates, while the microaggregates protect the SOC from microbial processes.

Because the SOM comprises a large and heterogeneous pool of C-enriched compounds, residence times of SOC in the organic pools range from a few minutes to hundreds of years. Residence times of relatively labile organic matter can be about 7 years in both silt and clay particles, whereas residence times for stable organics can reach ~400 years in silt and ~1000 years in clay (Buyanovsky *et al.*, 1994). Residence times of SOC in macro- and microaggregates may differ because of (1) differences in physiochemical attraction between mineral and organic particles and (2) location of the organic binding agents within the aggregates (Emerson, 1959). The SOC residence time depends on the geo-chemical composition (Greenland *et al.*, 1962), bonding agents (Tisdall and Oades, 1982), and size (Cambardella and Elliot, 1993; Six *et al.*, 2000) of aggregates. Residence time of C increases with decreasing aggregate size. Losses of SOC from macroaggregates are faster and larger than those from microaggregates (Besnard *et al.*, 1996) due to the differences in physical and chemical protection (Jastrow *et al.*, 1996).

The SOC protection is proportional to the specific external surface area of the clay particles, and to the monolayer interfaces between clay and sand particles. Emerson (1959) found that organic matter unavailable to microbial processes is confined between the clay crystals. The stabilizing power of clay is high, thus clay soils contain more SOC than sandy soils (Wild, 1988). Montmorillonitic clays store and protect SOC more than illitic and kaolinitic clays because montmorillonites possess greater surface area, more interlayer spaces, and higher swell and shrink potential than illites and kaolinites. Montmorillonites protect SOC by preventing the microbes from accessing the C-rich organic substrates, by controlling microbial population, and by preserving the microbial metabolites (Wild, 1988).

Although many studies indicate that all the SOC in microaggregates is biochemically recalcitrant with extremely low turnover rates and thus well protected (Tisdall and Oades, 1982; Wild, 1988), some studies, however, argue that SOC and microaggregate turnover can be more dynamic than has been reported (Golchin *et al.*, 1994; Jastrow *et al.*, 1996). The SOC physically is protected within microaggregates but chemically may be susceptible to microbial processes (Golchin *et al.*, 1994). The C associated with microaggregates can range from easily recognizable organic debris with rapid turnover rates to recalcitrant organics entrenched with clay particles with long residence times (Cambardella and Elliot, 1993; Jastrow *et al.*, 1996). Exact mechanisms of protection and resident times of SOC in macro- and microaggregates are not well understood (Cambardella and Elliot, 1993; Six *et al.*, 2000). Questions still remain on how the SOC flow is protected inside the microaggregates as a crucial mechanism for C sequestration (Angers and Chenu, 1998).

D. Distribution of Soil Organic Carbon in Aggregates

The distribution of SOC within the soil aggregates varies over time and space in interaction with management and soil type (Beare *et al.*, 1994a; Six *et al.*, 1999a; Table 4). The SOC concentration in macroaggregates under the same soil may vary depending on the soil management differences (Puget *et al.*, 1995; Figure 5). Beare *et al.* (1994a) showed that macroaggregates stored 61% of C in NT and 58% in CT soils. The authors also observed that macroaggregates stored 25% of POM in NT and only 20% in CT. The higher POM concentration in NT is due to the increased biological activity that incorporated more organic matter into the macroaggregates. Cultivation reduces the amount of POM in aggregates more than the mineral-associated organic fraction (Six *et al.*, 2000). Figure 5 shows that SOC within aggregates is usually higher in NT than in CT at the upper horizons, but it can be the opposite in subsurface horizons due to soil inversion in CT that tends to incorporate

TABLE 4
Distribution and protection of soil organic carbon (SOC) in aggregates

Location	Soil	Major conclusions	Reference
Australia	Fine sandy loam	Organic C is protected from microbial degradation within microaggregates but not in macroaggregates.	Adu and Oades (1978)
Australia	Fine sandy loam	Formation and stabilization of macro- and microaggregates depend on organic C concentration in the soil.	Tisdall and Oades (1982)
Sidney, NE	Loam	Concentration of organic C is greater in macroaggregates than in microaggregates.	Cambardella and Elliot (1993)
Athens, GA	Sandy clay loam	Microaggregates are formed in close interaction with particulate organic matter in the core of macroaggregates.	Beare *et al.* (1994a)
France	Silty clay	The SOC concentration increases with aggregate size, and organic C is younger in macroaggregates than in microaggregates.	Puget *et al.* (1995)
Batavia, IL	Silt loam	The SOC concentration in micro is lower than in macroaggregates, but the organic C in microaggregates is recalcitrant and well protected.	Jastrow *et al.* (1996)
Quebec, Canada	Silty loam	Young organic C accumulates in macroaggregates.	Angers and Giroux (1996)
Sidney, NE Wooster, OH Kellogg, MI Lexington, KY	Loam Silt loam Sandy loam Sandy clay loam	Intensive cultivation leads to formation of C-depleted microaggregates compared to no-till management.	Six *et al.* (1999a)
Akron, CO	Silt loam	Macroaggregate turnover reduces microaggregate formation and the protection of particulate organic matter.	Denef *et al.* (2001)
Athens, GA	Sandy clay loam	Microaggregates protect more SOC in no-till than in tilled soils.	Bossuyt *et al.* (2002)

residues at lower depths (Beare *et al.*, 1994b; Bossuyt *et al.*, 2002).

The distribution of SOC in aggregates also varies with textural classes (Greenland, 1971; Adu and Oades, 1978). Soil texture is often correlated with SOC concentration, with fine-textured soils having greater SOC (Hassink, 1992). Watson and Parsons (1974) found that silt stored 39% and clay 50% of the total SOC in loamy soils. In contrast, silt stored 47% and clay 36% of SOC in sand, sandy loam and calcareous clay, respectively. Christensen (1996) stated that <5 μm organo-mineral separates confine the highest organic C concentration, and that clay fraction comprises $>50\%$ and clay plus silt confine $>90\%$ of C concentration. Saggar *et al.* (1996) and Gonzalez and Laird (2003) also found that fine clay (<0.02 μm) had the highest and the coarse clay (0.2–2 μm) the lowest SOC concentration. Clay soils respond differently to SOC release because of their unique physical and chemical attributes (Six *et al.*, 2000; Chenu *et al.*, 2000). A full understanding of the processes involved in SOC distribution in aggregates cannot be complete, unless there is a recognition and thorough assessment of the complexity associated over time and space (Kooistra and van Noordwijk, 1996). Distribution of SOC within aggregates is unpredictable and heterogeneous due to its dynamic nature and management-induced mechanisms.

IV. ROLE OF AGGREGATE MECHANICAL PROPERTIES IN ORGANIC CARBON DYNAMICS

There exists a strong relationship between soil aggregation and SOC concentration. For example, SOC promotes soil aggregation, whereas aggregates in return store SOC, reducing the rate of SOM decomposition. Long-term studies have indicated that soil structural characteristics are positively correlated with SOC through a reciprocal relationship (Watts and Dexter, 1997). Six *et al*. (2000) found that losses in SOC were concomitant with degradation of soil structure. Studies on the effects of SOM on soil aggregation and structural stability are many (Golchin *et al*., 1995; Jastrow and Miller, 1998; Watts *et al*., 2001), but specific studies assessing the relationship between the mechanical properties of individual macro- and microaggregates and SOC concentration are limited (Zhang, 1994). The intrinsic mechanical behavior of individual aggregates may differ from that in the whole soil (Zhang, 1994) and most importantly can control the mechanical behavior of the bulk soil in relation to SOC sequestration.

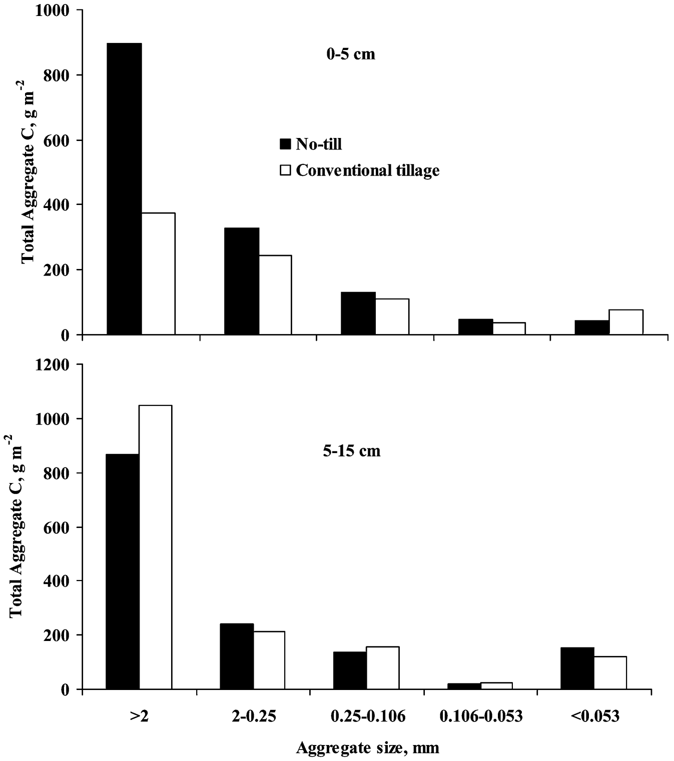

FIG. 5. Total aggregate C dynamics under no-till and conventional tillage management at two depth intervals (Drawn based on the data by Beare *et al.*, 1994a).

A. Density and Porosity of Aggregates

The density and porosity of aggregates are sensitive properties that may vary with SOC concentration (Loch, 2001). Increased SOC concentration is often correlated negatively with aggregate density and positively with aggregate porosity in the surface soils (Rogowski *et al.*, 1968; Poier and Richter, 1992). Zhang (1994) reported that aggregate porosity increased by ~25% when SOM increased by 8% while decreasing significantly the aggregate density. Similarly, aggregate density decreased with increasing SOC in soils under crop rotations (Hulugalle and Cooper, 1994). Increased crop residues in surface soils decreased the aggregate density compared to soils free from crop residues (Kay, 1998). Munkholm and Kay (2002) reported that aggregates between 4- and 8-mm had lower aggregate density and higher porosity in forage soils, but differences in soils under continuous corn were mixed. Therefore, density and porosity of aggregates can explain SOC dynamics, and information from these relationships can be used to assess SOC sequestration in the whole soil.

B. Soil Water Retention

Soil water retention characteristics of individual aggregates may also be a sensitive indicator of SOC concentration in aggregates (Barlow and Nash, 2002). Leffelaar (1993) observed that water retention is a valuable indicator of aggregate dynamics and vice versa. Yet, the effect of SOC on soil moisture retention of aggregates is little understood (Causarano, 1993). Snyder *et al.* (1993) observed that SOM increased with increasing moisture retention capacity both in the inter-aggregate pore space (0 to −0.033 MPa) and intra-aggregate porosity (−0.033 to −0.08 MPa). Soil moisture retention of < 8 mm aggregates increased with the incorporation of slightly decomposed SOM (Zhang, 1994). Inter-aggregate pore size affects soil moisture characteristic curve as well as in the encapsulation of organic C (Leffelaar, 1993). Emerson (1995) found that soil available moisture content in mesopores increased from 1 to 10 g of water per every 1 g of SOM added. Similarly, Watts and Dexter (1997) showed that for soil moisture potentials >−0.1 MPa, soils with higher SOC concentration have higher moisture content than soils with lower SOC concentration, indicating that increased SOC improves the retention of available water. While studying 60 different soils across the United States, Hudson (1994) observed that increased SOM usually results in increased moisture content at field capacity and wilting point. These studies show that soil moisture retention characteristics of aggregates can be sensitive to SOC dynamics, particularly at high energy levels.

C. Stability and Size of Aggregates

High SOC concentration commonly increases aggregate stability depending on the soil and site-specific factors affecting the aggregation. Chaney and Swift (1984) determined that aggregate stability and SOM ($r = 0.66$), total carbohydrate ($r = 0.71$), and humic material ($r = 0.60$) using 120 soils were significantly correlated. Similarly, Golchin *et al.* (1995) reported a strong correlation ($r = 0.86$) between POM and stability of 1–2 mm aggregates. Angers (1992) found, however, a low correlation between SOC and aggregate stability.

Correlations reported between aggregate sizes and SOC concentration are somewhat mixed (Table1). Jastrow *et al.* (1998) found low correlations between SOC and aggregate sizes as follows: $r = 0.43$ for >212 μm aggregates, 0.28 for >2000 μm aggregates, 0.54 for aggregates between 1000 and 2000 μm, and 0.02 for aggregates between 212 and 1000 μm. However, Hernanz *et al.* (2002) observed a positive correlation ($r = 0.62$) between 1–2 mm air-dried aggregates and SOC in a long-term study. Such contradictory results suggest that the processes of aggregate dynamics in interaction with SOC are complex and not well understood. Many underlying interactions and complex mechanisms among the binding agents are unaccounted for by a simple correlation. Tisdall and Oades (1982) attributed the low correlations to the following reasons: (i) only a fraction of the total SOM intervenes in aggregation, (ii) aggregation occurs up to a critical content of SOM beyond which increases in SOM are irrelevant, and (iii) aggregation is more related to free organic materials than to total SOM content.

D. Aggregate Strength

Correlation between tensile strength of aggregates and SOC concentration is often mixed. The SOC concentration may

(Rogowski *et al.*, 1968; Bartoli *et al.*, 1992) or may not (Causarano, 1993; Zhang, 1994; Watts and Dexter, 1997) be significantly correlated with aggregate strength. Aggregate strength tends to increase with decreasing SOC in silt loam soils (Watts and Dexter, 1997), but this trend depends on the interaction of soil properties with tillage-crop management practices (Beare *et al.*, 1994a). Aggregate strength increases in clayey soils but decreases in silt loam and sandy soils with increasing SOC (Kay, 1998). Changes in soil moisture content strongly affect the aggregate strength and SOC dynamics (Emerson, 1959; Causarano, 1993; Munkholm and Kay, 2002). Bonding strength of organic materials with mineral particles decreases with increasing soil moisture content, thus reducing the aggregate strength (Kay, 1998). The increase of SOC concentration may reduce the aggregate strength in wet but not in dry state in which clay content controls the mechanisms of aggregation (Greenland, 1971). Causarano (1993) observed that increased SOM strengthened wet aggregates and weakened the dry aggregates, but the correlations were low.

Existence of hierarchal architecture of soil aggregates is related to the aggregate strength. When an aggregate ruptures in a systematic order from macro- into microaggregates, it indicates that aggregate hierarchy exists. When an aggregate breaks down directly into silt and clay particles, it indicates that aggregate hierarchy does not exist (Tisdall and Oades, 1982). Planes of failure and weak spots are abundant in macroaggregates attributed to the presence of large pores and coarse organic residues (Oades and Waters, 1991). As a result, C-enriched macroaggregates tend to have lower aggregate strength than C-depleted macroaggregates, but further research is needed to elucidate the mixed findings of aggregate strength and SOC relationships.

V. CARBON SEQUESTRATION IN CLAY MINERALS

Current recommendations for enhancing C sequestration rely on the increased incorporation of plant residues and decreased intensity of tillage. Decreased soil disturbance leads to greater SOC storage (Lal *et al.*, 1998; Lal, 2002), but the SOC residence time is highly variable (Buyanovsky *et al.*, 1994). Partially processed organic C has a residence time between one month and one year, whereas residence times of organic C in clay minerals can be >100 years (Laird, 2001). Therefore, an understanding of the mechanisms of long-term C sequestration in clay minerals is important. Interaction of SOC with clay minerals is not only a prerequisite for C sequestration but for the stabilization of the aggregate architecture as a web for all the organo-mineral linkages (Emerson, 1954).

A. Mechanisms of Formation of Clay-Humic Complexes

Understanding of mechanisms of interaction between clay minerals and SOC involves the study of organo-mineral dynamics at a molecular scale (Table 5). Adsorption of organic molecules by clay minerals and intervention of electrostatic forces forms organo-mineral complexes with strong bonds between organic and clay particles (Emerson, 1959; Christensen, 1996). Organic anions of C-rich humic colloids and polysaccharides are adsorbed by clay particles through polyvalent metals (Ca^{+2}, Al^{+3}, and Fe^{+3}) following the basic microstructure: clay particle-polyvalent metal-organic matter (Greenland, 1965, 1971). One positive charge of the di- or tri-valent cation bonds the negative charge of the organic anion and the other charge bonds the negatively charged clay plate, thereby serving as a bridging mechanism. These anion-cation-anion complexes occur within clay domains forming clusters of microaggregates (Wild, 1988; Oades, 1984). The strong bonds between organic anion and clay crystals due to the action of the polyvalent cations prevent the organic materials from microbial decomposition. The strength of bonds depends on the valence of the metals bridging the inorganic and organic anions following this order: $Al^{+3} > Fe^{+3} > Ca^{+2} > Na^{+}$ (Tisdall, 1996). Formation of clay-humic complexes is further controlled by bonding mechanisms such as H-bonding, van der Waal's forces, and Coulombic attractions. Because of the strong bonds, higher energy expenditure is needed to rupture the silt- and clay-sized inter-particles in microaggregates than in sand particles (Edwards and Bremner, 1967). These organo-mineral complexes sequester C by means of three critical physical and biochemical functions (Laird *et al.*, 2001). First, organic materials react with clay particles through adsorption. Second, clay surfaces polymerize humic substances. Third, polymerized organic compounds are physically and chemically sequestered by clay crystals inaccessible to soil organisms.

Humic and fulvic acids are an essential component of organic complexes. Humic acids are particularly involved in aggregate stabilization, whereas fulvic acids are involved in chemical reactions within aggregates. Through organo-mineral complexation, they act as binding agents within the microaggregates (Tisdall and Oades, 1982). Detailed mechanisms of interaction between humic compounds and soil primary particles for C sequestration are not well understood, particularly with non-crystalline and amorphous organic compounds. Complex interactions among polysaccharides, humic and fulvic acids, and clay particles moderate soil structure and SOC dynamics (Greenland *et al.*, 1962; Tisdall and Oades, 1982).

B. Clay Mineralogy and C Sequestration

The importance of clay in retaining SOC across soils is influenced by mineralogical composition. Laird (2001) observed that the smectite-dominated clay favored a greater SOC storage than kaolinite- and chlorite-dominated clay minerals. The smectite-associated C is an important storage pool in microaggregates in 2:1 clay minerals. Differences in specific surface area may regulate clay dynamics for sequestering C. Saggar *et al.* (1996) showed that smectitic soils not only protect more SOC inside the clay crystals but the SOC turnover rates and residence times inside were longer than in other clay minerals due to increased surface area and retention locations (interstitial layers) of smectites. Gonzalez and Laird (2003) determined

TABLE 5
Interactions of organic molecules and clay minerals in soil organic carbon (SOC) sequestration

Location	Soil	Interactions	Reference
Australia	Fine sandy loam	C-enriched organic materials react with inorganic colloids and form clay-organic complexes.	Greenland (1965)
England	Clay	Polyvalent cations adsorb organic C molecules to clay surfaces.	Wild (1988)
Canada	Silt loam	Organic C increases with clay content. Clay fraction stores ∼60% of SOC.	Gregorich *et al.* (1988)
Australia	Clay	Polymers and polysaccharides link clay crystals forming organo-mineral complexes.	Emerson *et al.* (1986)
Canada	Silty clay loam and clay loam	C-enriched humic and fulvic acids are found in coarse clay, whereas aliphatics are in clays and noncrystalline minerals.	Schnitzer and Kodama (1992)
Australia	Clay	Cations bridge clay minerals and organic molecules, thus controlling microaggregation.	Oades (1984)
Canada	Silty clay loam and clay loam	Organic C is positively correlated with extractable Al and chloritized vermiculite but negatively with total clay content.	Monreal and Kodama (1997)
New Zealand	Clay	Surface area and electrical charge of clay minerals rather than total clay content control the protection of SOC from microbial decay.	Saggar *et al.* (1996)
Waseca, MI	Clay loam	Dense humic compounds associate with coarse and medium clay particles, whereas diffuse humic compounds interact with fine clay minerals.	Laird *et al.* (2001)
Germany	Fine sand	Organic C is bound to the clay matrix in interaction with hydrous Al and Fe minerals particularly in subsurface horizons.	Kaiser *et al.* (2002)
Ames, IA	Silt loam	Clay mineralogy controls the formation and stabilization of C-enriched humic compounds, which are primarily concentrated on fine smectite clay surfaces.	Gonzalez and Laird (2003)

that C-enriched humic compounds were preferentially attached to the surface of smectitic minerals. However, Wattel-Koekkoek *et al*. (2001) found that accumulation of organic C was independent of clay mineralogy except that organic C in kaolinite was rich in polysaccharides whereas that in smectite was abundant in aromatic compounds, implying that binding mechanisms of clay and organic C interactions vary among clay types. Monreal and Kodama (1997) observed that SOC was positively correlated with extractable Al and Fe, chloritized vermiculite, and mica but was negatively with the total clay content, indicating that consideration of clay type is important in addition to total clay content for addressing C sequestration in clay minerals.

Clay minerals vary tremendously in the ability to bind organic molecules. The rate of C sequestration depends on the surface area, ionic charge, type, and chemical and geo-chemical composition of clay minerals. Different clay minerals retain different kinds of organic compounds due not only to differences in binding mechanisms but specific soil and environmental conditions. The biochemistry of clay-organic complexes can be different from the whole soil and can greatly control the magnitude and rate of C sequestration (Laird, 2001; Kaiser *et al.*, 2002). Indeed, Laird (2001) stated that humic compounds are closely associated with clay minerals and together develop unique physical and chemical characteristics differing from the whole soil in terms of C sequestration dynamics. Clays protect the soil biomass and integrate the metabolic products of the soil biota with the soil matrix (Saggar *et al.*, 1999). Nature and size of macro- and microaggregates depend on the type of clay, polyvalent metal, and organic compounds (Tisdall and Oades, 1982).

VI. STATISTICAL FUNCTIONS OF SOIL AGGREGATE AND SOIL ORGANIC CARBON RELATIONSHIPS

Many simple and sophisticated statistical tools are available to assess soil aggregate and SOC relationships including pedotransfer functions (PF), principal component analysis (PCA), and multivariate analysis (MA). The PF are potential tools to translate data into variables that are usually expensive to measure. The term PF was initially introduced as a predictive function of soil hydraulic properties (Bouma, 1989), but can also be useful to assess the sensitivity of soil structure to changes in SOC and to integrate with other soil properties. The PF approach is mostly used as a tool to translate readily measurable soil properties into variables needed in predictive models. However, a limitation of the use of PF is the large number of undefined variables. These functions are often soil and site-specific and need the inclusion of many variables, which diminishes their utility for interpolating results to all soils. Formulation of PF requires long-term data from soils managed under the same parent material and climatic characteristics. Although PF has long been used for exploring interrelationships among variables (Minasny and McBratney, 2002), it has not been widely used for studying soil structure and SOC relationships. The PCA involving a transformation of SOC correlated variables into uncorrelated variables (principal components) may be another useful approach (Shinjo *et al*., 2000). The PCA is useful to detect, identify, and classify meaningful variables related to SOC storage and to minimize the dimensionality of the data collected (Martin *et al*., 2002). The MA is another statistical approach that permits the study of multiple response variables as affected by SOC dynamics (Zanini *et al*., 1998; Brown *et al*., 2000). Use of advanced statistical tools such as PCA and MA for assessing SOC distribution in aggregates is not well known (Eghball *et al*., 1994).

VII. ASSESSMENT METHODS OF SOIL ORGANIC CARBON DISTRIBUTION IN AGGREGATES

Data on SOC distribution within aggregates are needed to affect a better understanding of aggregate dynamics for SOC sequestration, but measurements often require complex and expensive procedures. Whereas methods for quantifying total organic C in biological materials are abundant and well developed, those for assessing distribution of SOC within the aggregates are relatively few (Table 6). The SOC distribution within aggregates is heterogeneous, and its assessment is a function of the method used (Kooistra and van Noordwijk, 1996). Therefore, attention to methods is important to draw valuable conclusions of the interrelationships between soil structure and SOC.

A. Classical Approaches

Traditional techniques of dry and wet sieving of aggregates are severely constrained in the quantification of C in different size fractions. Sieving and pretreatments procedures performed prior to the determination of aggregate size distribution affect the SOC distribution in aggregates (Gale *et al*., 2000b). For example, rapid wet sieving can cause significant disruption of aggregates due to internal air pressure differences and impact the microbial community and dynamics responsible for enhancing aggregation (Sainju *et al*., 2003). Unstable macroaggregates

TABLE 6

Techniques used in recent studies for assessment of aggregate dynamics in relation to soil organic carbon (SOC) storage

Location	Soil	Determination	Methods	Reference
Germany	Fine sand	Particle size distribution in microaggregates	X-ray attenuation	Leifeld and Kögel-Knabner (2003)
Fort Valley, GA	Loamy fine sand	Aggregate size Total C	Dry sieving C analyzer	Sainju *et al.* (2003)
Germany	Fine sand	^{14}C Clay mineralogy	Mass spectrometer Electron microscopy	Kaiser *et al.* (2002)
Athens, GA	Silty clay loam	Aggregate size ^{14}C	Wet sieving Oxidixer and Liquid Scintillation Analyzer	Bossuyt *et al.* (2002)
Denmark	Sandy loam	Total C Aggregate strength Soil strength	C analyzer Crushing test Shear vane	Munkholm and Kay (2002)
Akron, CO	Silt loam	Aggregate size Total C ^{13}C	Dry sieving C analyzer Mass spectrometer	Denef *et al.* (2001)
Iowa	Silt loam	Aggregate size ^{13}C	Wet sieving Oxidixer and liquid Scintillation Analyzer	Gale *et al.* (2000a, 2000b)
Sidney, NE	Loam	Aggregate size C analyzer ^{13}C	Dry sieving Total C Mass spectrometer	Six *et al.* (2000)
Germany	Fine sand and clay	Isolation of organo-mineral complexes	Ultrasonic dispersion	Schmidt *et al.* (1999)
Sidney, NE	Loam	Mineral associated soil organic matter	Hexametaphosphate for aggregate dispersion	Cambardella and Elliot (1993)

fail to withstand these induced forces as compared to stable macroaggregates, affecting the assessment of C distribution. Dry sieving is a preferable technique over wet sieving for maintaining a relatively intact habitat and activity of soil microorganisms (Schutter and Dick, 2002), which, however, reflects dry field conditions with no rain or irrigation. Results comparing the sieving (wet and dry) and pretreatments methods are mixed (Cambardella and Elliot, 1993; Gale *et al.*, 2000b).

Several techniques have been used since the 1970s for assessing SOC distribution within aggregates (Table 6). Many have used chemical extractants and physical separations to isolate and quantify SOC in microaggregates (Jenkinson, 1971). Ultrasonic vibration is another technique used to isolate organo-mineral complexes from bulk soils, and to study the interactions between the inorganic and organic components of aggregates (Edwards and Bremer, 1967). Most methods are questionable because they have not fully unraveled the SOC dynamics at a molecular scale to relate to C sequestration at regional and global scales.

B. Contemporary Approaches

Since the 1990s researchers have explored a variety of techniques to study the mechanisms of SOC storage in aggregates (Table 6). Many researchers have used a combination of physical and chemical procedures (Elliot and Cambardella, 1991; Cambardella and Elliot, 1992). Cambardella and Elliot (1993) assessed the SOC distribution by dispersing the aggregates with sodium hexametaphosphate and sieving the dispersed fractions through 53-μm sieve to remove sand and POM components and to measure the total organic C. These procedures allow the study of SOC associated with mineral particles in aggregate fractions (Beare *et al.*, 1994a).

A relatively new method consists in tracing the fate of organic inputs in aggregates by analyzing stable-isotope of ^{13}C. This method has been widely used for quantifying the turnover of organic matter in plants (Martin *et al.*, 1990). The basic principle of this method is that concentration changes of ^{13}C natural isotope induced by the cultivation of plant species different from previous species are monitored. For example, C rates from C-3 plants developed under native prairie vegetation are compared with that of C-4 plants by isotopic analysis (Bender, 1968; Balesdent *et al.*, 1987). The analysis consists of obtaining C isotopic ratios of ^{13}C/^{12}C from automated measurements on CO_2 coupled with isotope mass spectrometer (Balesdent *et al.*, 1987).

Some researchers have used the radioactive C isotope (^{14}C) analysis to determine the turnover time and distribution of SOC pools in aggregates (Adu and Oades, 1978; Gale and Cambardella, 2000). The basis for this approach is that complex interactions of cosmic rays housed in the atmosphere generates natural radiocarbon (^{14}C) and integrates with the CO_2 molecules. Plants have the ability to absorb ^{14}C from the atmosphere through photosynthesis and store it until senesce. Thus, the ^{14}C concentration in soil is correlated with that in living plants. The ^{14}C concentration decreases rapidly with vegetation loss (Adu and Oades, 1978). Combining C-labeling approaches with nondegradative physical and chemical fractionation techniques can be a better approach to studying the SOC distribution in aggregates.

Examination of fractal dimension of soil macro- and microaggregates using X-ray computed tomography (CT) is another approach for studying aggregate dynamics in relation to SOC distribution. The computer imaging can characterize the macropore networks from a 3-dimensional perspective in aggregates (Perret *et al.*, 2003). This approach permits the internal visualization of attributes of soil aggregates to store SOC, and can further our understanding of the mechanisms of formation and connectivity of soil pores in aggregates and structural configuration of the macro- and microaggregates. The X-ray computed tomography has been extensively used to examine soil structural attributes (Crawford *et al.*, 1995) but seldom to address SOC distribution in aggregates.

Most of the current methods determine the SOC in bulk soil aggregates. Physical separation methods, which permit the removal of different concentric layers forming aggregates, can be an improved approach to evaluate the SOC distribution. Santos *et al.* (1997) developed an apparatus, which enclosed in an erosion chamber, permits the examination of cross sectional layers of individual aggregates. The authors showed that the device improved the physical separation of ion gradients in aggregates and facilitated the identification of physical and chemical processes in aggregation. The apparatus can be adapted to a systematic study of SOC concentration gradients within aggregate layers in combination with C isotope techniques and ultrasonic procedures. Schmidt *et al.* (1999) and Leifeld and Kögel-Knabner (2003) used an ultrasonic dispersion procedure to separate organo-mineral compounds within soil aggregates. Advanced techniques including X-Ray Scattering and Scanning Electron Microscopy are additional methods that can offer new insights into the surface area and geo-chemical attributes of aggregates while examining the SOC distribution within the aggregates (Laird *et al.*, 2001; Gonzalez and Laird, 2003). Scanning electron microscopy is highly useful to detect SOC differences in the soil profile and to visualize organo-mineral interactions and dynamics (Kaiser *et al.*, 2002). These techniques would be extended to fully visualize external and internal attributes of macro- and microaggregates including recalcitrant molecules binding organo-mineral complexes, specific surface area, pore space geometries, morphology, microporosity, mineralogy, and access pathways to external soil microorganisms into the core of micro- and macroaggregates. Until now, despite the abundance of techniques of SOC determination in aggregates, SOC turnover times, distribution and dynamics of input and output rates of SOC, particularly within the microaggregates, are still unanswered (Wang and Hsieh, 2002).

VIII. RESEARCH NEEDS

There are several knowledge gaps in understanding the interactions of soil structure and SOC. Current state-of-knowledge has not unraveled completely the underlying complex processes involved in the sequestration, stability, dynamics, and residence times of SOC in macro- and microaggregates. Some of the major constraints are:

1. Although studies on the effects of plant residues and soil management on SOM dynamics are numerous, there is still much uncertainty in the dynamics of organic binding agents in the macro- and microaggregation (Gale *et al.*, 2000a). For example, surface-derived residues appear to behave differently than the root-derived residues in aggregation (Gale *et al.*, 2000b). A better understanding of the distribution and confinement of microbially derived SOM within aggregates would allow a better assessment and comprehension of the SOC sequestration in the whole soil.
2. There is some contradiction among the available models in explaining the aggregate architecture in relation to the SOC dynamics. Classical models state that primary particles form microaggregates and then macroaggregates (Tisdall and Oades, 1982). Contemporary models, in contrast, show that macroaggregates are formed first and then release microaggregates (Oades and Waters, 1991; Six *et al.*, 2000). Thus, there is a need to develop a unique conceptual model of aggregate hierarchy that describes all the fundamental mechanisms of aggregate dynamics in relation to SOC storage.
3. Formation of clay-organic complexes has been widely studied (Edwards and Bremner, 1967; Tisdall, 1996), but few studies have integrated such information to explain the exact physical and chemical mechanisms of SOC sequestration in clay minerals (Gonzalez and Laird, 2003). Biochemistry of clay minerals differs from the whole soil for the formation of C-enriched humic compounds in clay fractions (Laird *et al.*, 2001). Understanding of the processes of long-term SOC sequestration in clay minerals can help implement proper management systems to form clay-organic complexes.
4. Studies on the effects of tillage and crop management on SOM and soil physical and chemical properties are abundant (Cambardella and Elliot, 1993; Puget *et al.*, 1995; Lehmann *et al.*, 2001), but there is not enough information that relates SOC dynamics to mechanical properties of individual macro- and microaggregates such as pore size distribution, Atterberg limits, shear strength, tensile strength, density, and compressibility in different soils under long-term tillage and crop management systems. Specific studies assessing the relationship of mechanical properties of aggregates to SOC concentration are few (Causarano, 1993; Watts and Dexter, 1997). Mechanical behavior of individual aggregates may not only differ from that of the bulk soil but also can determine the behavior of the whole soil system in relation to long-term SOC sequestration.
5. Current methods to assess SOC distribution in aggregates have not completely unraveled the physical, chemical, and biological mechanisms that control the interactions of soil structure versus SOC distribution. The use of C isotope techniques has advanced our understanding of aggregate structure and SOC relationships, but comparative studies using more advanced techniques are needed to further examine the SOC dynamics, particularly within microaggregates at a molecular level (Kaiser *et al.*, 2002).
6. A systematic assessment of the mechanisms of SOC retention in soil aggregates requires a comprehensive and integrated approach of modeling and research experimentation. Modeling could provide insights on the type of soil management practices that can be implemented to enhance C sequestration under different soil and climate settings. Models that incorporate data on SOC distribution in macro- and microaggregates to predict soil potential for storing atmospheric C are desirable.

IX. CONCLUSIONS

Plants and SOM control soil structure and SOC dynamics. The quantity and quality of residues determine the formation and stabilization of aggregate structure for SOC sequestration. Plant residues through microbial processes generate complex substances that serve as a mechanical framework for linking soil particles into aggregates. Plant roots and residues are the primary organic skeleton to enmesh the inorganic particles together and build macro- and microaggregates while sequestering SOC. There are three major organic binding agents of aggregation: temporary (plant roots, fungal hyphae, and bacterial cells), transient (polysaccharides), and persistent (humic compounds and polymers).

Natural and agricultural ecosystems play an important role in the conversion of atmospheric CO_2 into SOM pool to sequester SOC. Conversion of natural ecosystems into agricultural lands for intensive cultivation severely depletes SOC pools. A judicious management of soils under competing and diverse land uses is the key to increasing SOM. Use of crop rotations and diverse cropping systems combined with NT practices followed by proper fertilization and irrigation can enhance SOC sequestration.

Classical models of soil aggregation state that the aggregate organization involves organic binding of primary soil particles into microaggregates where these microaggregates encapsulate SOC and then form macroaggregates. Modern models instead propose that microaggregates occluding SOC are actually formed within the macroaggregates rather than vice versa. Microaggregates in interaction with decomposed organic materials develop within macroaggregates before releasing new microaggregates for the next cycle of macroaggregation. Both models recognize that C-enriched fresh plant residues are responsible for the formation and stabilization of macroaggregates. Mechanisms of SOC and aggregate dynamics are complex and embrace a range of organic binding processes associated with the architectural organization of macro- and microaggregates in interaction with time and space.

A relevant mechanism of SOC storage in aggregates is the sequestration of plant debris in the core of soil microaggregates inaccessible to microbial processes. This mechanism is essential to stabilizing aggregates and sequestering SOC. Magnitude of SOC sequestration in the soil system depends on the residence time of SOC in aggregates. Microaggregates are bound to old organic C, whereas macroaggregates contain younger organic material. The SOC confinement in the interior of microaggregates is the source for long-term C sequestration in terrestrial systems. Ultimately, interactions of clay minerals with C-enriched humic compounds control the protection, residence times, and turnover of SOC within the microaggregates.

Many techniques have been used to assess the SOC distribution in aggregates. Classical methods include SOC determination in aggregate fractions by wet and dry sieving of bulk soil. Isotopic methods including the determination of ^{13}C and ^{14}C with mass spectrometry are techniques to quantify the turnover and storage of organic materials in soil aggregates. Other techniques involve the use of computed tomography, X-ray scattering, and X-ray microscopy to examine the internal porosity and inter-aggregate attributes of macro- and microaggregates. The latter approaches may improve our knowledge about the interactions of SOC and aggregate structure, and the location or distribution of SOC within the matrix of aggregates.

Although studies on soil management influences on soil aggregation are numerous, research on SOC distribution in aggregates and internal biophysical attributes of aggregates promoting C sequestration is scarce. Questions still remain on how SOC interacts physically and chemically with aggregates for increasing soil potential for sequestering C. Future research must enhance our ability to understand the mechanisms responsible for the dynamics of aggregates in correlation with SOC and their importance and functions to sequester C.

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