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Increased Soil Aggregate Stability by Altering Contents and Chemical Composition of Organic Carbon Fractions via Seven Years of Manure Addition in Mollisols

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Abstract: Mollisols include an abundance of soil organic carbon (SOC) which is easily influenced by fertilization management. Manure addition could enhance soil aggregate stability; however, the dominating factor affecting its stabilization remains controversial. The fertilization practices were initiated in 2012 to investigate the influences of different fertilization managements on the contents and molecular characterization of organic carbon (OC) fractions, and to clarify the underlying mechanism of soil aggregate stability change. NoF (non-fertilizer), CF (only chemical fertilizer), CF + DM (chemical fertilizer plus single dairy manure at 15 t ha^{-1}), and CF + 2DM (chemical fertilizer plus double dairy manure at 30 t ha^{-1}) treatments were established. This research was aimed at exploring the potential mechanism that affects aggregate stability in Mollisols through the variation of contents and chemical composition of OC fractions, and screening out the appropriate fertilization practice on promoting SOC stabilization and crop yield under 7-year manure addition. Compared to CF, 7-year manure addition significantly enhanced SOC content by 17.4–35.9% at 0–10 cm depth, which was evidenced from the contribution of increased aromatic compounds with 4.3–19.9%. Simultaneously, compared with CF, CF + DM and CF + 2DM both significantly enhanced dissolved organic carbon and easily oxidizable organic carbon contents by 12.5–37.7% at a 0–30 cm soil layer. In regard to soil aggregates, the increased OC content and mass percentage of macroaggregates, and the decreased mass percentage of free microaggregates both improved aggregate stability under manure addition at 0-30 cm soil layer, which was proven to be the increment in mean weight diameter (MWD) and geometric mean diameter (GMD) values by 17.6–22.1%. Moreover, CF + DM and CF + 2DM raised aromatic compound amounts of POM fractions within macroaggregates [M(c)POM] by 5.6–11.6% and within free microaggregates (Fm-POM) by 4.3–10%. Furthermore, CF + DM and CF + 2DM both significantly increased maize yield by 5.7% and 4.2% compared to CF, but no significant difference was observed between CF + DM and CF + 2DM treatments. Collectively, physical protection through the occlusion within aggregates of POM might be the central mechanism for soil aggregate stability of manure addition in Mollisols. The manure addition of 15 t ha^{-1} was the effective management method to enhance SOC stabilization and crop yield in Mollisols.

Keywords: molecular characterization; particulate organic matter; silt and clay fractions; Fourier transform infrared spectroscopy; aromatic components; physical protection

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1. Introduction

Soils, the biggest carbon (C) reservoir in the terrestrial ecosystems globally, stores over three- and two-fold C levels than the global terrestrial plants and atmosphere, respectively [1]. Soil organic carbon (SOC) can have a critical impact on improving soil structure

(especially increasing soil aggregate stability), promoting soil quality, enhancing crop productivity, and moderating climate change [2–5]. Increased C inputs derived from manures, composts, and crop residues can increase the equilibrium level of SOC, thus increasing SOC storage in arable soils [6]. The availability of manure addition in elevating SOC content hinges on the type of manure, the addition rate, and the application duration [7–9].

Nearly 4000 years ago, the practice of manure application in agricultural ecosystems was initiated in China [10], whereas the remarkable increase in crop yield and economic benefits in the past two decades from synthetic fertilizers caused reduced interest in manure addition to the soils [11]. In China, the annual production in farmyard manure is over four billion tons, but the percentage that is applied to the agricultural soils is very small because of its high consumption in transporting, processing, and spreading [12]. Synthetic (chemical) fertilizers may increase soil fertility and crop yield when managed appropriately [13]. However, the overuse of synthetic fertilizers for a long time may result in soil nutrient loss, soil acidification, and structural deterioration which reduce SOC stability and crop productivity [14,15]. Multiple long-term soil advantages associated with organic carriers of plant nutrients make manure application a logical choice for crop growth where manure is available.

OC composition in agricultural soils is uneven, complex, and dynamic due to it being spatially heterogeneous [16]. The variation of SOC in response to fertilizer practices has been attributed to a series of soil and management conditions [17]. Labile OC components are more sensitive and responsive early indicators of changes in agricultural practices owing to its readily decomposable and fast turnover times, which promotes nutrient cycling and biological activity [18]. Some labile OC fractions (e.g., easily oxidizable OC (EOC), dissolved OC (DOC), and microbial biomass C (MBC)) are considered important indicators for assessing SOC stocks associated with different soil managements [19].

SOC addition stimulates water stable aggregate (WSA) formation through cementation, reorganization, and flocculation of primary mineral particles [20]. Soil aggregation is usually regulated by cementing agents (e.g. plant residues, polysaccharides, and fungal hyphae), physical (occlusion within aggregates), and mineral mechanisms (reactive cations bridge) [21]. Macroaggregates (>0.25 mm) supply the least physical protection in terms of organic matter are structurally susceptible to agricultural measures such as fertilization and tillage [22]. However, microaggregates (<0.25 mm) are crucial to long-term soil organic matter maintenance due to less exposure to microorganisms [23]. The physical-chemical combined grouping method can more deeply and comprehensively explore the soil aggregate stability mechanism from the organic matter components distributed in intra-aggregates of different particle sizes [24]. The responses of synthetic fertilizer and manure addition in OC content within macroaggregates and microaggregates, and soil aggregate stability are inconformity [25]. For example, 21-year synthetic fertilizer application with or without organic manure neither influenced soil aggregate stability nor aggregate-associated SOC under wheat-maize rotation on calcareous soils because of the similar mineralization rates from the different hierarchically organized aggregates [26]. The results of our previous research illustrated that, compared to continuous maize, soil aggregate stability was enhanced by a 41.8–58.2% increment in mean weight diameter (MWD) and geometric mean diameter (GMD) by chemical fertilizer application under maize-soybean and fallow-soybean rotations in Mollisols [2]. Consequently, soil aggregation is a continuous and complex process, and thus the apparent discrepancies in aggregate stability and aggregate-associated OC in the different fertilization practices may depend on soil type, experimental duration, climate condition, and cropping systems [11].

Mollisols play a crucial part worldwide in maintaining sustainable agricultural development and ecological security. However, worldwide, Mollisols have experienced a rapid growth in intense cultivation due to large-scale agricultural expansion over the past few decades, resulting in the dramatic decline in natural fertility and SOC content [27]. Previous research demonstrated that 2-year manure addition obviously enhanced SOC content at 0–10 cm, and enhanced MBC and DOC contents at 0–20 cm soil depth under a maize-

soybean rotation in bulk Mollisols, compared to no fertilization and chemical fertilizer application alone [28]. The 6-year consecutive addition of cattle manure significantly enhanced SOC stability on account of the increased mass percentage of macroaggregates, and reduced mass percentage of microaggregates and non-aggregated silt and clay fractions [29], whereas limit information is obtainable in regard to the effect of manure application on the characteristics of various OC components, particularly the molecular structure in intra-aggregates by physical-chemical combined grouping at different soil depths in Mollisols. Thereby, the purposes of this study were: (1) to explore the influences of 7-year manure addition on the contents and chemical composition of OC fractions; (2) to probe into the potential mechanism affecting the soil aggregate stability; and (3) to select the appropriate fertilization practice on promoting SOC stabilization and crop yield under a maize-soybean rotation in Mollisols.

2. Materials and Methods

2.1. Experimental Platform

A long-term located experiment was set up at the Hailun Station of Soil Erosion Monitoring and Research (126°49′ E, 47°21′ N) in the Northeast Institute of Geography and Agroecology, Chinese Academy of Sciences. The study area belongs to the semi-humid continental monsoon climate. The mean annual precipitation is 553.9 mm, and the mean annual cumulative temperature is 2450 °C. This soil type is known as typical Mollisols [30] and its fundamental physical and chemical properties have been presented by Zhou et al. [31].

This experiment was initiated in the spring of 2012 with four different fertilization practices under the soybean (*Glycine max* (Merrill.) L.)-maize (*Zea mays* L.) rotation. The maize belonged to the spring maize. Soybeans were initially planted in May 2012 and subsequently spring maize was planted in May 2013, followed by the same rotation sequence. In the experimental plot, randomized block distribution was adopted with three replications for each treatment. The length, width, and row spacing of each plot was 8 m, 5.36 m, and 0.7 m, respectively. At the beginning of May each year, the crops began to grow. Soybean and maize crops used the local staple varieties, and the plant density of soybeans and maize were 240,000 ha^{-1} and 48,000 ha^{-1}, respectively. The herbicides of acetochlor with 1500 mL ha^{-1} and thifensulfuron-methyl of 120 g ha^{-1} were immediately applied for weed control after the crops were planted.

The fertilization measures included NoF (non-fertilizer), CF (only chemical fertilizer), CF + DM (chemical fertilizer plus single dairy manure at 15 t ha^{-1}), and CF + 2DM (chemical fertilizer plus double dairy manure at 30 t ha^{-1}). For dairy manure application, the decomposed manure by fermentation naturally with moisture content of 10% was applied into 30 cm of soil depth after the crops were harvested in October. The nutrient content on the dry weight of manure was as follows: total C of 454 g kg^{-1}, total N of 20.7 g kg^{-1}, total phosphorus (P) of 7.93 g kg^{-1}, and total potassium (K) of 11.8 g kg^{-1} [32].

In terms of soybeans, urea, diammonium phosphate [$(NH_4)_2HPO_4$], and potassium sulfate (K_2SO_4) were used as N, P, and K of 20.3 kg ha^{-1}, 21.2 kg ha^{-1}, and 2.2 kg ha^{-1}, respectively, which was applied as a base fertilizer at the seeding stage. With regard to maize, urea, $(NH_4)_2HPO_4$, and K_2SO_4 were used as N, P, and K of 69 kg ha^{-1}, 28.1 kg ha^{-1}, and 6.1 kg ha^{-1}, respectively, which were applied as base fertilizers at the seeding stage. Simultaneously, the 69 kg ha^{-1} of N that was used as urea was added to the maize fields in mid-June.

2.2. Soil Sampling

After the crops were harvested in October 2019, the soil samples for 0–10 cm, 10–20 cm, and 20–30 cm depths were gathered. Both soybeans and maize had been planted for 4 years. Three soil samples from each plot were randomly taken by soil-drilling and then mixed into one single sample. Following this, the soil samples were separated into three parts. Two parts were air-dried, ground, and finally passed through the 0.25 mm and 2 mm sieves.

Next, every sample was homogenized on account of coning and quartering methods [33] to measure soil aggregates with 2 mm sieve, and SOC and EOC contents with 0.25 mm sieve. The other part of the soil samples was disrupted and separated by hand, passed through the 2 mm sieve, and finally kept at 4°C for the measurement of DOC and MBC contents.

2.3. Procedure of Soil Aggregate Classification

The wet-sieving method was adopted to separate the soil aggregates [34]. 50 g air-dried soil samples were distributed evenly onto the two nested sieves (0.25 mm and 0.053 mm) in order to fractionate three aggregate size fractions. The nest was held at the highest point of the oscillating cylinder and the distilled water was added into each cylinder until the water was above the topmost sieve. Before the wet-sieving action, the soils were submerged into water for a period of 10 min. During the whole process, the oscillation time of 10 min, stroke length in the vertical direction of 4 cm, and the frequency of 900 cycle h^{-1} always remained constant. Firstly, the soil aggregates were initially divided into three fractions of "macroaggregates" (M), "free microaggregates" (Fm), and "non-aggregated silt + clay" (nA-MOM) according to the above procedures.

Secondly, the macroaggregates were saturated by distilled water for 10 min and put into the two sieves (0.25 mm and 0.053 mm) fixed on the reciprocating shaker with 186 cycle min^{-1}. This operation obtained the fractions of "coarse POM within macroaggragates" [M(c)POM] and "microaggregates-within-macroaggregates" (mM) staying in the 0.25 mm and 0.053 mm sieves, respectively. The fraction in the rinse water was called "silt + clay fractions within macroaggregates" (M-MOM).

Finally, the fraction of Fm was inverted in the 1.7 g cm^{-3} NaI heavy solution. Through centrifugation, the light fraction of "fine POM within free microaggregates" [Free(f)POM] was separated out. Next, the heavy fraction was acquired by dispersion with the $(NaPO_3)_6$ solution of 0.5 mol L^{-1} and passing through the sieve of 0.053 mm. The substance of "POM from heavy fraction within free microaggregates" (Fm-POM) was preserved on the 0.053 mm sieve, and the fraction of "silt + clay fractions from heavy fraction within free microaggregates" (Fm-MOM) existed in the rinse water. Simultaneously, the same procedure was applied to separate the mM substance into three fractions which were named as "fine POM within mM" [M(f)POM], "POM from heavy fraction within mM" (mM-POM), and "silt + clay fractions from heavy fraction within mM" (mM-MOM). The residual substance on each sieve was collected, dried at 60–80 °C, and the weight was calculated.

The schematic diagram of aggregate classification was presented in Figure 1, and the abbreviations and full names of organic matter and OC fractions, and soil aggregate stability indicators mentioned in the current research were described in Table 1.

Table 1. The abbreviations and full names of various organic matters and organic carbon fractions, and soil aggregate stability indicators.

Abbreviations	Full Names
SOC	soil organic carbon
EOC	easily oxidizable organic carbon
DOC	dissolved organic carbon
MBC	microbial organic carbon
M	macroaggregates
Fm	free microaggregates
nA-MOM	non-aggregated silt + clay fractions
mM	microaggregates within macroaggregates
POM	particulate organic matter
M(c)POM	coarse POM within macroaggregates
M(f)POM	fine POM within mM
mM-POM	POM from heavy fraction within mM

Table 1. *Cont.*

Abbreviations	Full Names
Free(f)POM	fine POM within free microaaggregates
Fm-POM	POM from heavy fraction within free microaggregates
M-MOM	silt + clay fractions within macroaggregates
mM-MOM	silt + clay fractions from heavy fraction within mM
Fm-MOM	silt + clay fractions from heavy fraction within free microaggregates
MWD	mean weight diameter
GMD	geometric mean diameter
D	fractal dimension

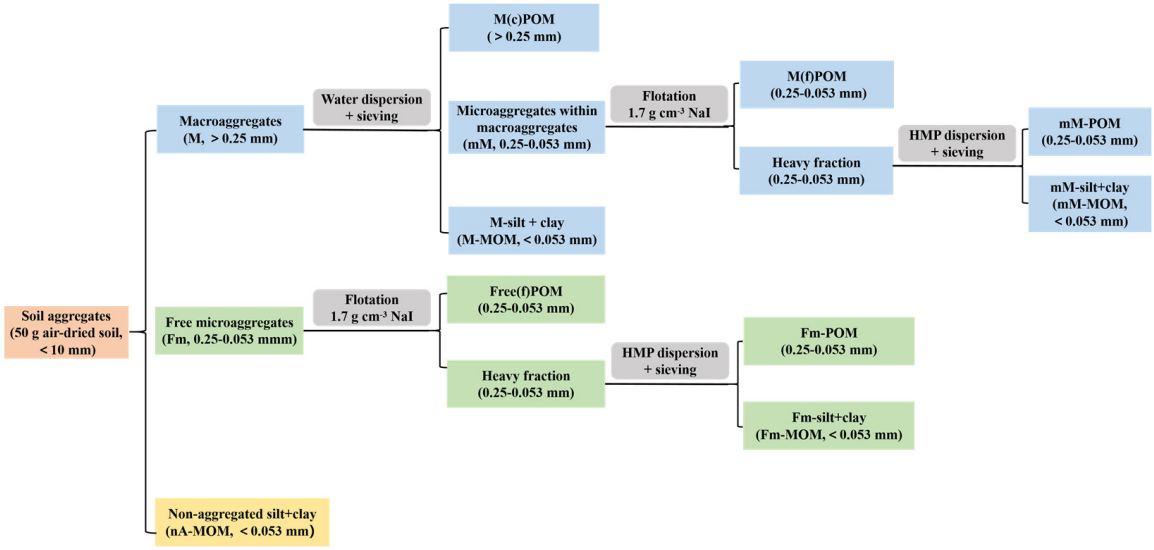


Figure 1. The procedure of soil aggregate classification. HMP, sodium hexametaphosphate. The specific meanings of organic matter fractions with abbreviations are presented in Table 1.

2.4. Measurements of Soil Chemicals and Physical Properties

Bulk soils refer to intact soils before grouping. Soil total carbon (STC) in bulk soils was measured by an element analyser (FlashEA 1112, Thermo Finnigan, Rodano, Italy). STC content is equivalent to the SOC content because the inorganic carbon content was very low and could be ignored in Mollisols [35]. EOC was determined by the oxidation method of 333 mmol L^{-1} $KMnO_4$, and MBC was performed by the fumigation technology of $CHCl_3$ [36,37]. Then, the OC content of each fraction was measured through the above element analyser. The detailed determination methods of soil pH value, bulk density, total N, NH_4^+ -N, NO_3^--N, olsen-P, available K, and microbial biomass nitrogen (MBN) were performed according to the research by Zhou et al. [3].

2.5. Data Calculation

The indicators of soil aggregate stability include MWD, GMD, and fractal dimension (D). These three indexes were calculated by Equations (1)–(3) based on the methods described by Bavel, Kemper and Chepil, and Yang et al., respectively [38–40].

$$MWD = \sum_{i=1}^{n} (\overline{Ai}Bi) \quad (1)$$

$$GMD = \exp\left(\frac{\sum_{i=1}^{n} Bilg\overline{Ai}}{\sum_{i=1}^{n} Bi}\right) \quad (2)$$

$$(3 - D)lg\left(\frac{\overline{Ai}}{Amax}\right) = lg\left(\frac{C_{(\delta \leq \overline{Ai})}}{C}\right) \quad (3)$$

where Bi represents the weight proportion occurring in the corresponding size fraction, $\overline{Ai}$ means the mean diameter of each size (mm), determined by the mean-value of the minimum and maximum diameter in each aggregate, Amax represents soil particles diameter with 2 mm, and $C_{(\delta \leq \overline{Ai})}$ indicates the total mass of diameter $\leq \overline{Ai}$.

2.6. Fourier Transform Infrared Spectroscopy Analysis

The Fourier transform infrared spectroscopy (FTIR) spectrum was analysed using the spectrometer (Nicolet IS10, Thermo Fisher, Waltham, MA, USA). Firstly, the soil sample was put in the 60–70 °C drying oven for 4 h to make the tablet and in order to protect it from interference from the hydroxyl group. Secondly, the electronic equilibrium with the accuracy of 1/10,000 was used to weight 200 mg dried KBr with spectral purity and 1 mg dried soil sample (KBr:sample = 200:1 ratio). Thirdly, the above substances were ground to powders of <2 μm particle size by the agate mortar and were then mixed well. Fourthly, the mixed materials were pressed into a thin slice with 10 t cm^{-2} pressure, and held for 1 min. Finally, the materials began to scan within the scanning range, number, and the resolution of 4000–400 cm^{-1}, 32 times, and 4 cm^{-1}, respectively. During scanning, the spectrum of background air was automatically subtracted.

2.7. Statistical Analysis

The SOC contents (g C kg^{-1} fraction) in all organic matter components including aggregate grouping, macroaggregates and free microaggregates in physical-chemical combined grouping directly measured by the element analyser were all converted to the SOC content of each component in the whole soil (g C kg^{-1} soil), and the percentage of each component in the total soil mass was used for conversion analysis. The graphs were performed through Origin 7.5 (OriginLab Corporation, Northampton, MA, USA). The semi-quantitative FTIR peak area analyses were constructed by integration via OMNIC 8.2 (Thermo Nicolet Corporation, Madison, WI, USA). Regression analysis and Pearson correlation analysis (PCA) were both conducted through SPSS 20.0 (International Business Machines, Co., Ltd., Armonk, NY, USA), and the structural equation model was performed using SPSS Amos 21.0 (International Business Machines, Co., Ltd., Armonk, NY, USA).

3. Results

3.1. The Contents of SOC and Labile OC Fractions in Bulk Mollisols

With respect to bulk soils, compared with either NoF or CF treatment, CF + DM and CF + 2DM treatments significantly ($p < 0.05$) enhanced SOC content with 17.4–35.9% at 0–10 cm soil depth (Figure 2). The SOC content at 10–20 cm soil layer was increased ($p < 0.05$) by 11.9% and 7.3% under CF + 2DM treatment compared to NoF and CF treatments, respectively, whereas the treatment effect on SOC content was not observed at a 20–30 cm depth.

In regard to labile OC fraction contents, CF + DM and CF + 2DM treatments significantly ($p < 0.05$) raised EOC content compared to NoF (by 34.1–45.7%) and CF (by 24.8–37.7%) across the three soil depths (Figure 2). At the same time, DOC content under CF + DM and CF + 2DM treatments were also 18.1–24% higher than that of NoF, and 12.5–18.3% higher than that of CF among all three depths. CF + DM and CF + 2DM treatments enhanced ($p < 0.05$) MBC content by 19.9% and 14.9% at 0–10 cm; by 19.8% and 14.3% at 10–20 cm; and by 12.6% and 8.2% at 20–30 cm soil depth, respectively. In addition, compared with CF, CF + DM and CF + 2DM treatments both increased MBC content by 19.2% and 14.2% at 0–10 cm soil depth.

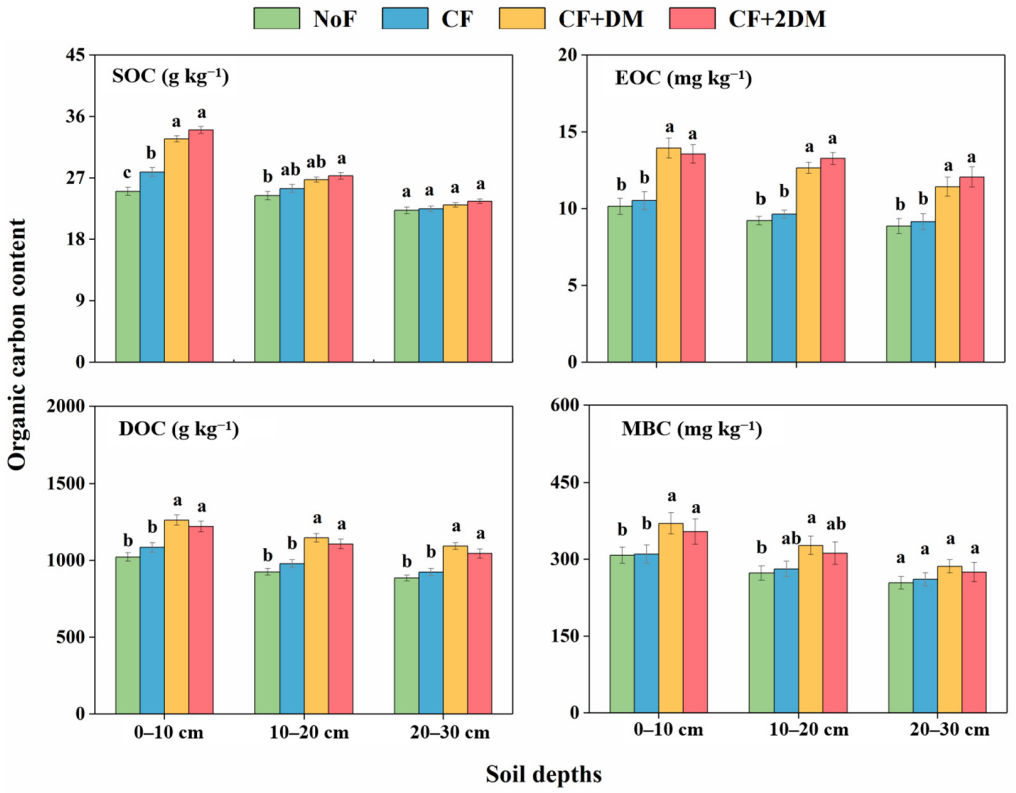


Figure 2. The contents of soil organic carbon and organic carbon fraction in bulk Mollisols under different fertilization treatments. In the same soil layer, the various letters of the same fraction exhibited significant differences at the $p < 0.05$ level via Tukey's HSD test under the four treatments. NoF, non-fertilizer. CF, only chemical fertilizer. CF + DM, chemical fertilizer plus with single dairy manure. CF + 2DM, chemical fertilizer plus with double dairy manure. The specific meanings of organic carbon fractions with abbreviations are presented in Table 1.

3.2. The Contents of OC Fractions within Soil Aggregates and Soil Aggregate Stability

Across the four treatments and three depths, the largest OC content was presented in the macroaggregates of 11.05–20.90 g kg^{-1}, and next was free microaggregates of 6.19–6.81 g kg^{-1}. nA-MOM fractions of 3.05–3.55 g kg^{-1} demonstrated the minimum OC content (Figure 3). In addition, the Fm-MOM, mM-MOM, and M-MOM fractions illustrated the biggest OC contents in free microaggregates, microaggregates within macroaggregates, and macroaggregates.

The effects of various OC fractions within aggregates under different fertilization managements were demonstrated in Figure 3. Macroaggregates-associated OC content under CF + DM and CF + 2DM treatments were 50.1% and 53.8% higher ($p < 0.05$) than that of NoF, and 29.0% and 32.2% higher ($p < 0.05$) than that of CF at 0–10 cm depth. CF + DM and CF + 2DM treatments all significantly enhanced SOC content of M(c)POM by two-fold compared to NoF, and by 82.4% and 83.5% compared to CF at 0–10 cm soil layer. Simultaneously, in comparison with CF treatment, the OC content of M(f)POM under CF + DM and CF + 2DM treatments was 2.9% and 4.3% higher ($p < 0.05$) at 0–10 cm, and 10.5% and 14.0% higher significantly at 10–20 cm soil layers.

Moreover, compared to NoF treatment, CF + DM and CF + 2DM treatments raised ($p < 0.05$) the OC content of Free(f)POM by 16.7–53.1% across all three soil depths (Figure 3). Simultaneously, compared to CF treatment, the OC content of Free(f)POM was also increased by 16.7% and 26.7% under CF + DM and CF + 2DM treatments at 20–30 cm soil depth. However, the increase in amplitude of 10.9% and 11.4% at 0–10 cm and 10–20 cm soil depths was only presented in the CF + 2DM treatment. CF + DM and CF + 2DM treatments averagely increased OC content of Fm-POM by 2.51% and 4.05% than

that of NoF treatment, and by 1.66% and 3.19% than that of the CF treatment among all three depths, whereas these four fertilization practices demonstrated no significant effect on the OC contents of Fm, nA-MOM, mM, M-MOM, and Fm-MOM fractions at any soil depth (Figure 3 and Figure S1).

The MWD and GMD values were higher, but D was lower under manure addition (CF + DM and CF + 2DM treatments) than NoF or CF treatment, regardless of soil depth (Figure 3). Specifically, compared to CF, CF + DM and CF + 2DM treatments significantly ($p < 0.05$) and averagely enhanced MWD by 20.5% and 22.1%, and raised GMD by 17.6% and 18.5%, but decreased ($p < 0.05$) D value by 2.30% and 2.31% across the three soil depths, respectively.

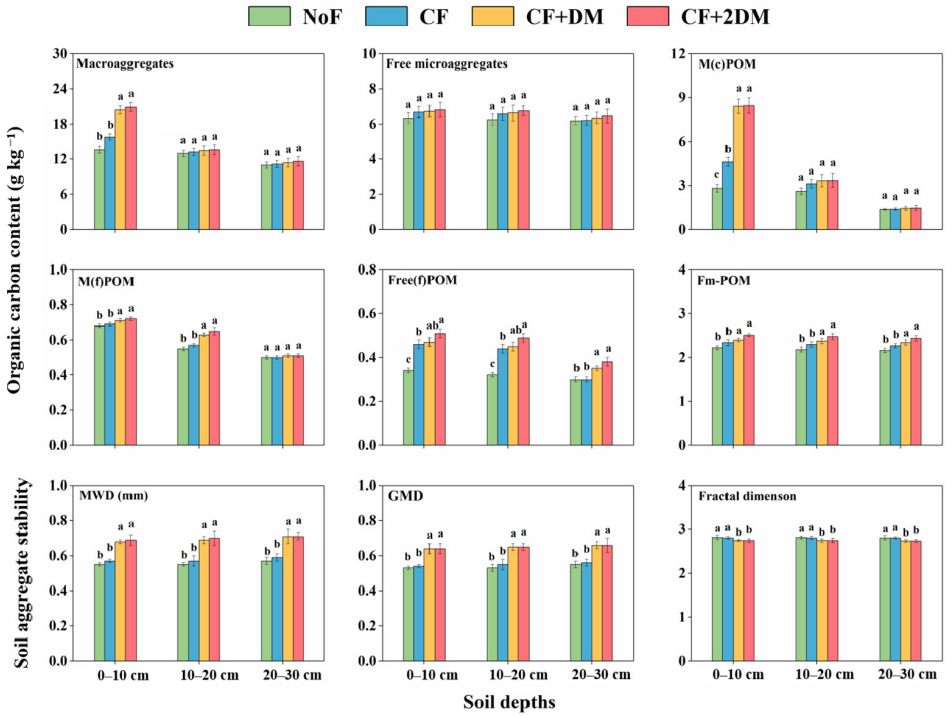


Figure 3. The contents of organic carbon fractions within hierarchical aggregates and soil aggregate stability under different fertilization managements. In the same soil layer, the various letters of the same fraction exhibited significant differences at the $p < 0.05$ level via Tukey's HSD test under the four treatments. NoF, non-fertilizer. CF, only chemical fertilizer. CF + DM, chemical fertilizer plus with single dairy manure. CF + 2DM, chemical fertilizer plus with double dairy manure. The specific meanings of organic matter fractions with abbreviations are presented in Table 1.

3.3. Size Distribution within Soil Aggregates

Across all fertilization treatments, macroaggregates showed the biggest mass percentages (44.8–52%), the following was free microaggregates (23.2–30.7%) and non-aggregated silt and clay fractions (23.1–25.5%) (Figure 4). Moreover, the greatest mass percentages of free microaggregates, microaggregates within macroaggregates, and macroaggregates was presented in Fm-MOM, mM-MOM, and M-MOM fractions, respectively, (Figure 4 and Figure S2).

As the amount of dairy manure increased, the proportion of macroaggregates in any soil depth gradually increased, while the proportion of free microaggregates gradually decreased, and the proportion of nA-MOM remained unchanged (Figure 4). Compared to CF, the proportion of macroaggregates was significantly ($p < 0.05$) raised by 7.2–10.6% across the three soil depths under manure addition. However, compared with CF, CF + DM and CF + 2DM, treatments significantly reduced the proportion of free microaggregates by 13.1–15.2% across the three soil depths.

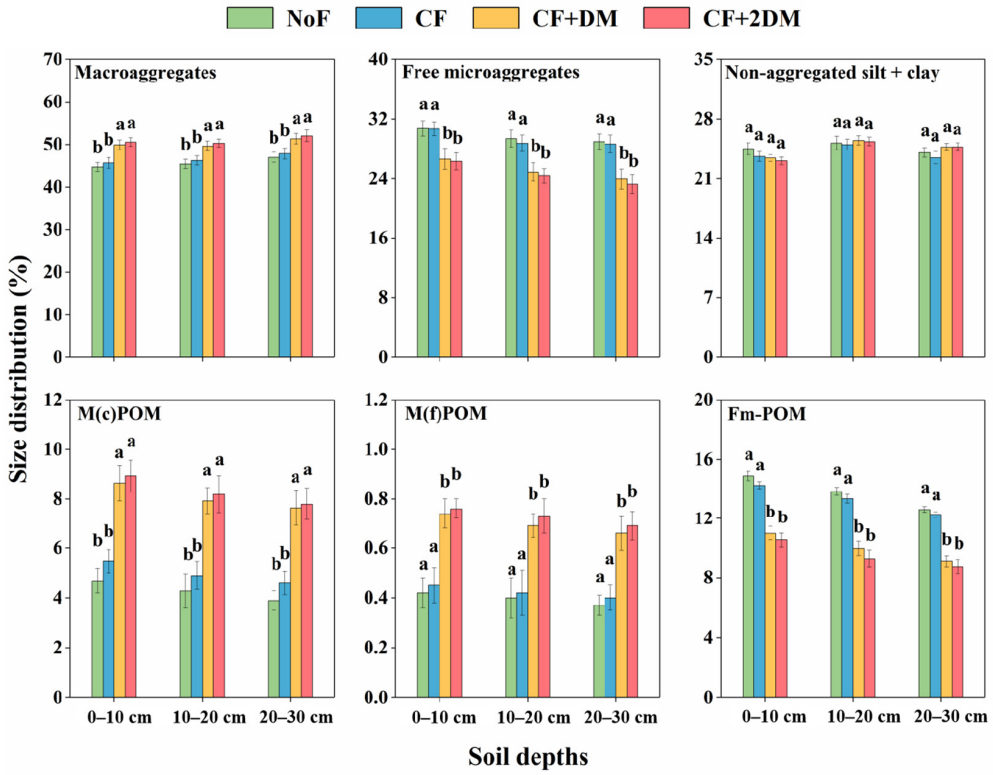


Figure 4. Size distribution within hierarchical aggregates under the four fertilization managements. In the same soil layer, the various letters of the same fraction exhibited significant differences at the $p < 0.05$ level via Tukey's HSD test under the four treatments. NoF, non-fertilizer. CF, only chemical fertilizer. CF + DM, chemical fertilizer plus with single dairy manure. CF + 2DM, chemical fertilizer plus with double dairy manure. The specific meanings of organic matter fractions with abbreviations are presented in Table 1.

Moreover, the proportions of M(c)POM and M(f)POM were both significantly enhanced by 58.2–69.7% under CF + DM and by 64.3–73.8% under CF + 2DM compared to CF treatment across the three soil layers, respectively (Figure 4). In addition, the proportion of Fm-POM was 22.8–30.5% lower under manure addition than CF among the three depths. Whereas, the proportion of mM, mM-POM, Free(f)POM, mM-MOM, M-MOM, and Fm-MOM was not influenced by the manure addition at any soil depth (Supplementary Figure S2).

3.4. Chemical Composition in Various OC Fractions on Account of FTIR Spectroscopy

The FTIR spectrum in bulk Mollisols and aggregates exhibited the existence of alcohol and phenol substances of 3623 cm^{-1}, aliphatic substances of 2920 cm^{-1}, aromatic substances of 1620 cm^{-1}, and carbohydrates of 1030 cm^{-1} (Supplementary Figures S3–S6). Among the four treatments and three soil layers, the amounts of carbohydrates (75.9–86%) was the biggest for any OC fraction, subsequently by alcohol and phenol (7.1–17%), aromatic (4.1–6.5%) and aliphatic (2.0–2.6%) substances (Supplementary Tables S1–S4).

Simultaneously, as the amount of dairy manure increased, alcohol and phenol materials showed a tendency to decrease, while aliphatic and aromatic substances, and the 1620/2920 rate presented a tendency to increase. However, the carbohydrate substances remained constant regardless of treatments and OC fractions (Supplementary Tables S1–S4). To be specific, compared to CF, CF + DM and CF + 2DM significantly enhanced the relative absorption peak area of aromatic compounds by 4.3–19.9% and the 1620/2920 rate by 1.1–13.5% among the three soil depths in all measured OC fractions.

3.5. Crop Yield

The order of single grain weight, single core weight, single gross weight, and crop yield in maize were CF + DM > CF + 2DM > CF > NoF, but no significant differences were exhibited between CF + DM and CF + 2DM treatments (Figure 5). Specifically, CF + DM and CF + 2DM treatments significantly increased the single grain weight by 4.5% and 3.6% and enhanced single gross weights by 3.9% and 3.2% compared to the CF treatment. Simultaneously, compared with NoF treatment, CF, CF + DM, and CF + 2DM treatments significantly ($p < 0.05$) raised crop yield by 2.5%, 8.1%, and 6.3%, respectively. Furthermore, CF + DM and CF + 2DM treatments also enhanced ($p < 0.05$) crop yield by 5.7% and 4.2% than that of CF treatment.

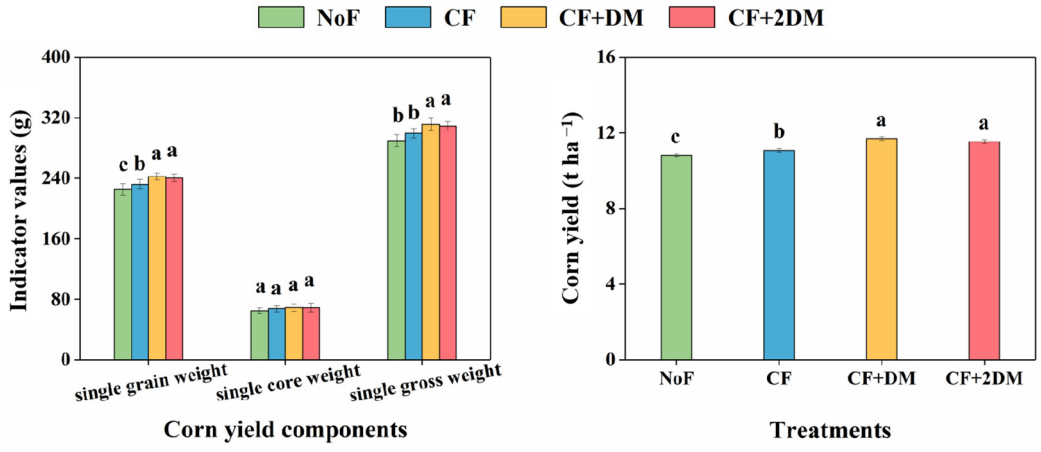


Figure 5. Responses of corn yield components to different fertilization managements in 2019. In the same soil layer, the various letters of the same fraction exhibited significant differences at the $p < 0.05$ level via Tukey's HSD test under the four treatments. NoF, non-fertilizer. CF, only chemical fertilizer. CF + DM, chemical fertilizer plus with single dairy manure. CF + 2DM, chemical fertilizer plus with double dairy manure.

3.6. PCA and Correlation Analysis

Regardless of fertilization treatments and soil depths, PCA illustrated that the CF + DM treatment was the most closely relevant to the aromatic substances amounts and 1620/2920 rate in bulk soils; macroaggregates, free microaggregates, nA-MOM fractions; and M(c)POM, mM-MOM, Fm-POM and Fm-MOM fractions (Figure 6).

The MWD and GMD were both significantly ($p < 0.05$) and positively correlated to SOC and EOC contents and the OC content of Fm-POM (Figure 7). Simultaneously, mass macroaggregates and mass macroaggregates/mass microaggregates ratio were positively correlated with MWD and GMD values, but they were negatively correlated to D value.

Macroaggregates were negatively and significantly ($p < 0.01$) correlated with the relative absorption peak area of alcohols and phenols (3623 cm^{-1}), aliphatics (2920 cm^{-1}), aromatics (1620 cm^{-1}), carbohydrates (1030 cm^{-1}), and the ratio of 1620/2920 (Figure 8). Similarly, the OC content of nA-MOM, Fm-POM, and Fm-MOM fractions were also significantly ($p < 0.01$) and negatively related to the relative absorption peak area of 3623 cm^{-1}. However, a significant ($p < 0.05$) and positive correlation was obtained between the OC content of a nA-MOM fraction and the relative absorption peak area of 2920 cm^{-1}. Simultaneously, positive and significant ($p < 0.01$) correlations were also presented between the OC content of Fm, nA-MOM, Fm-POM, Fm-MOM fractions and the relative absorption peak area of 1620 cm^{-1}. In addition, the SOC content in bulk soils and the OC content of M-MOM fractions presented a positive and significant ($p < 0.05$) relationship with the relative absorption peak area of 1030 cm^{-1}, but the OC content of Fm, nA-MOM, mM, Fm-POM and Fm-MOM fractions showed the opposite relationship with the relative absorption peak area of 1030 cm^{-1}. Except for macroaggregates, the other fractions were all positively and significantly ($p < 0.01$) correlated to the 1620/2920 rate.

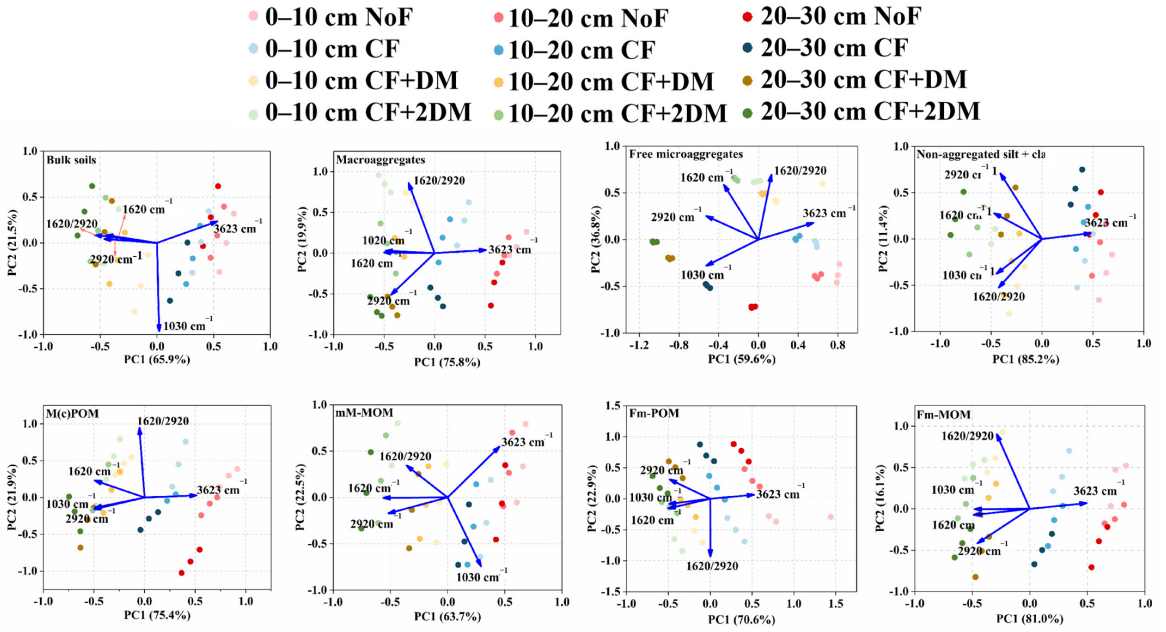


Figure 6. Principal component analysis from the relative absorption peak area and ratio of functional groups by FTIR technology. NoF, non-fertilizer. CF, only chemical fertilizer. CF + DM, chemical fertilizer plus with single dairy manure. CF + 2DM, chemical fertilizer plus with double dairy manure. The specific meanings of organic matter fractions with abbreviations are presented in Table 1.

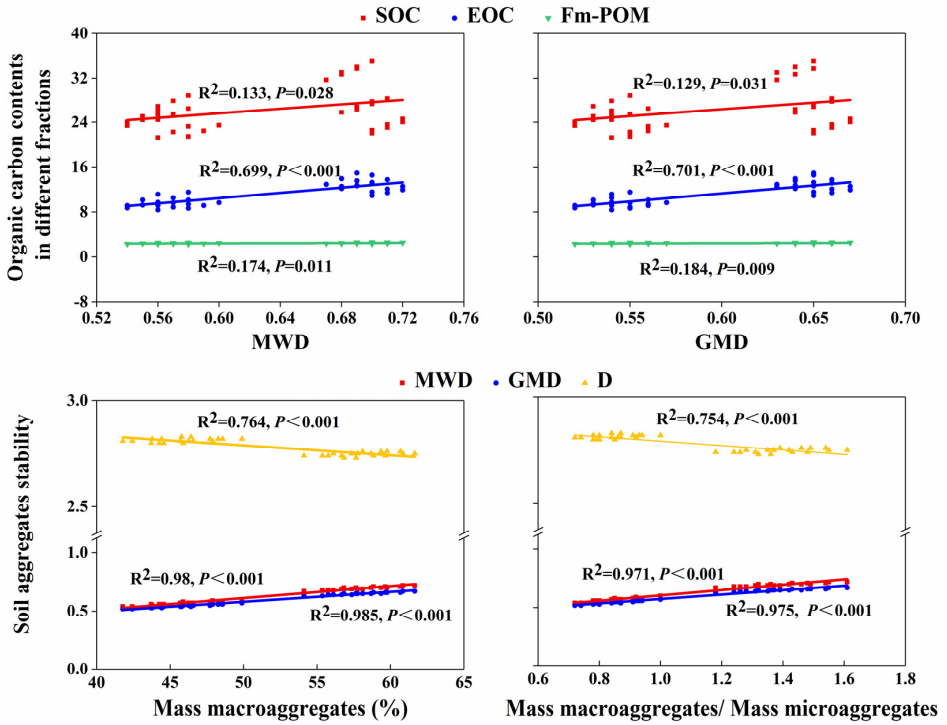


Figure 7. Linear regression equations of the measured parameters. Mass macroaggregates and mass microaggregates represent the proportion of mass distribution in soil aggregates within macroaggregates and free microaggregates, respectively, n = 36. The units of SOC, EOC, OC content of Fm-POM, and MWD are g kg^{-1}, mg kg^{-1}, g kg^{-1}, and mm respectively. The specific meanings of organic matter and organic carbon fractions with abbreviations are presented in Table 1.

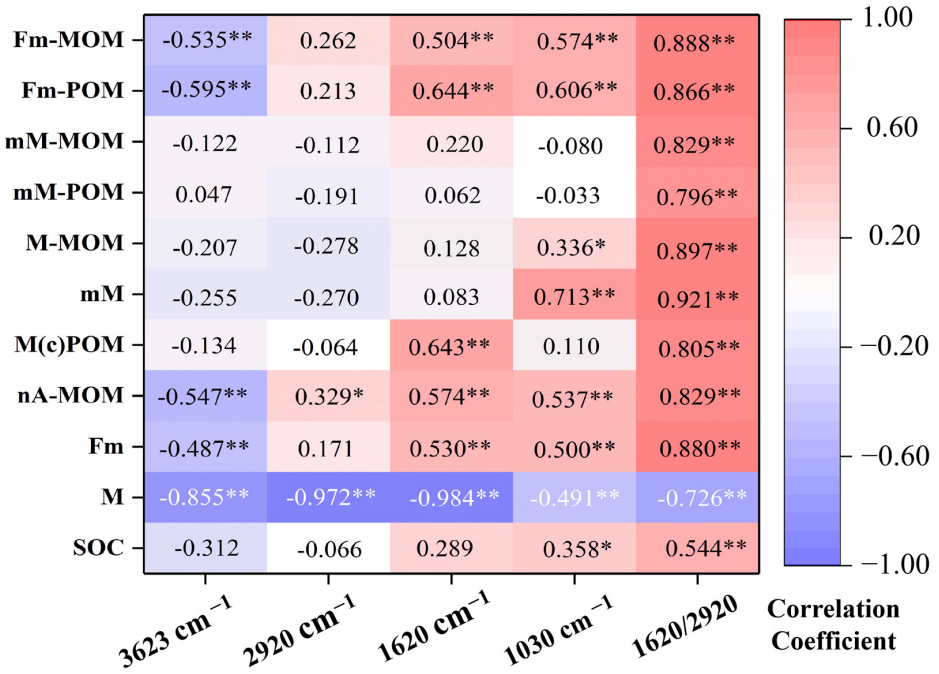


Figure 8. Correlation analysis between the relative area of infrared absorption peak and organic carbon content in bulk Mollisols and aggregates sizes. * $p < 0.05$, ** $p < 0.01$, $n = 36$. The specific meanings of organic matter and organic carbon fractions with abbreviations are presented in Table 1.

4. Discussion

4.1. Fertilization Practices Related to Variation in the Contents of SOC and Labile OC Fractions

The significantly increased SOC content under manure addition at 0–20 cm soil depth of bulk soils in the present research might be due to the increased hydrolysable and mineralizable C supply that promoted the activity of decomposing microorganisms [41], which is in line with previous research results [42].

Simultaneously, the increased DOC, EOC, and MBC contents at 0–30 cm under 7-year manure addition might be ascribed to the incremental increases of fresh organic matter by the application of organic fertilizer. Research suggests that part of the applied organic fertilizer is used to renew the cell components of the microorganism itself, and the other part is oxidized to the energy substance required by the activity of the microorganism itself [43]. In addition, the significant and positive correlation between SOC and labile OC fractions (EOC, DOC, and MBC) contents in our study illustrates that labile SOC content is closely related to TOC content to a large extent. Liang et al. indicated that MBC was significantly related to the contents of DOC, which implied microorganisms might use the dissolution of DOC to satisfy their own growth and reproduction [44].

Furthermore, though SOC and the content of labile OC fractions increased with the addition of manure application, no significant difference was observed between single- and double-rate manure, which indicates that SOC and the content of labile OC fractions reach the threshold after 7-year continuous application of high-rate manure (30 t ha^{-1}). Li et al. observed a similar phenomenon: net primary and soil C saturation were increased by appropriate nitrogen addition, but higher nitrogen application rates decreased DOC content on account of the incremental C consumption by soil microbes [11,18]. Additionally, higher manure addition rates would generate environmental pollution of greenhouse gas emissions and nutrient leaching due to the increased concentrations in mineral N, and available P and K [45]. It was worth noting that higher manure application rates were not practicable for smallholder farmers because of the low economic benefits [11].

4.2. Responses of Fertilization on OC Fractions within Soil Aggregates and Aggregate Stability

The current research illustrates that the soil under manure application included a higher mass percentage of macroaggregates, which presented a better soil structure. The proportion of different aggregate sizes verified that manure leads to nA-MOM fractions (smaller aggregates) gathered into larger aggregates, partly because of organic materials, addition-induced cementing agents that promote soil agglomeration and structural stabilization [46].

M(f)POM, M(c)POM, Free(f)POM, and Fm-POM fractions under manure application contributed more to smaller aggregates and aggregation in the present study. Previous research reported that mM-MOM and Fm-POM could be used as the cementing agents for stable macroaggregates and microaggregates formation, respectively [47,48]. However, the combined application of chemical fertilizer and manure could not obviously impact the proportion of aggregate sizes in calcareous soils because of the similar mineralization rates in different aggregate sizes [26].

The previous research proposed that the amount of each soil aggregate size class mainly controlled the OC content stored in different size fractions [49]. On the basis of the aggregate hierarchy model, macroaggregates were formed by microaggregates through the organic cementation. At the same time, macroaggregates were broken into microaggregates, the two were mutually reinforcing, thus macroaggregates played a pivotal part in the storage and sequestration of SOC [50]. The OC content decreased with the declined soil aggregate size in the present research which was identical to the results of Guan et al. [24]. Therefore, the soil aggregate hierarchy was an increment of OC content with the growing size class in essence [51].

Additionally, the current research exhibited that M-MOM, Fm-MOM, and mM-MOM fractions contained more OC content than POM in both macroaggregates and free microaggregates, indicating that OC in macroaggregates and microaggregates was mainly protected by the inorganic binding of silt and clay-sized fractions in Mollisols. Analogously, Smith et al. and Guan et al. both suggested that the vital material in the formation and stabilization of soil macroaggregates was MOM rather than POM in highly weathered tropical soils and fluvo-aquic paddy soils, respectively [24,52].

The aggregate distribution partly determined aggregate stability [53]. Macroaggregates included more plant-derived (i.e., new) C than other soil aggregates size classes, and were, in reverse, less stable because macroaggregates were formed by microaggregates through plant-derived organic matter [50,54]. The current research found that manure addition had the obvious response on macroaggregate-associated OC at 0–10 cm depth, while no significant influence was presented at 10–30 cm soil depth. The lack of change might be because of the balance between SOC outputs and inputs where SOC loss was offset by litter C inputs when manure was applied, indicating that manure addition impeded SOC accumulation at a 10–30 cm soil depth in Mollisols. However, the OC content of Free(f)POM inside free microaggregates was significantly raised across the 0–30 cm soil layer, which might be because of the transfer with a large amount of mobile DOC in microaggregates from the topsoil to the lower soil under the manure application, and more DOC from the topsoil might promote soil microbial activity and function, therefore DOC was converted into microbial residual C which could be deposited in POM [55].

Our findings showed that the OC content of mineral-associated organic matter (nA-MOM, Fm-MOM, M-MOM, and mM-MOM) all did not change under manure addition. The SOC sequestration of silt and clay fractions might be due to the combination of organic minerals and physical protection, rather than the stoichiometry of nutrient elements [54]. Our previous findings demonstrated that the N, P, N/P ratio and OC content of MOM were always logarithmic, which indicates that the SOC sequestration nutrient status threshold of the MOM component had been reached under 7-year manure addition, and the SOC sequestration was dependent on the physical binding properties of MOM [55].

Regardless of application rates, compared with chemical fertilizer application alone, manure addition positively affected soil aggregate stability according to the significant

increase in MWD and GMD values, and the significant decrease in D value across 0–30 cm of soil depth in our findings. The increased aggregate stability resulted in an obvious decrement in the mass percentage of nA-MOM, and OC was physically inaccessible to soil microbes, causing macroaggregates-associated OC to be raised [56]. The increase in aggregate stability might be because of the changes in the quantity and quality of litter returned through the addition of organic materials. Firstly, the high input of a litter originating from manure stimulated soil wetting and promoted soil aggregation, thus causing the soil aggregate stability to enhance [57]. Secondly, the increment of hydrophobic organic matter components could improve soil water-aggregate stability [58]. Consequently, a significant increase in hydrophobic aliphatic and aromatic functional groups, and a significant decrease in hydrophilic alcohol and phenol functional groups could, in part explain the increase of soil aggregate stability. Bronick and Lal also showed that the increase in soil aggregation depended on phenol substances [20]. Thereby, we infer that SOM quality was closely correlated with the biochemical composition of manure applied to the soil, and the relationship between substrate quality and soil aggregate stability warrants further study.

Linear regression equations illustrate that the soil aggregate stability enhanced with the increment in SOC and EOC contents, the OC content of Fm-POM, mass macroaggregates, and mass macroaggregates/mass microaggregates ratio in our findings. Thus, these measured parameters were the sensitive and crucial indicators for the aggregate stability under long-term manure application in Mollisols.

Fm-POM fraction was a labile source in soil free microaggregates and was strongly impacted by the application of organic materials in recent decades [59]. Our results corroborated Bhogal et al. and Yague et al. in their statements that POM fraction was an early indicator of long-term influences under soil quality management [60,61]. Current research distinguished it from others works in which aggregate stability was related to only SOC in bulk soils [62,63]. Due to its rapid turnover and low specific density, the POM fraction was considered as the decomposing plant and manure part, stimulating transient binding agents of polysaccharide production through microbial activities. The continued polysaccharide production would generate increases in soil aggregate stability [47,64,65].

4.3. Fertilization Practices Induced Differences in Infrared Spectroscopy

The significant increased amounts of aromatic compounds in bulk soils and all fractions under manure addition demonstrates that these fractions were developing towards aromatization and complication with organic materials application, and thus promoted C sequestration. Fresh organic materials applied to the soil made microorganisms preferentially decompose the relatively unstable and large-sized materials. As time increased, the microorganisms might have adapted to this easily decomposable material (that is biocompatibility), causing SOM to reach a saturation phenomenon [66]. Then, the decomposition of microorganisms would be weakened, resulting in a weakening of the degradation of lignin-like substances [67]. Therefore, 7-year manure addition will make most of SOC exist in the stable form.

Macroaggregates were mainly derived from the O-alkyl C groups of plant residues and were more easily decomposed and utilized by plants and microorganisms. Microaggregates were mainly composed of aromatic C, carboxyl C, and alkyl C groups derived from the metabolites of microbial life activities, and thus high stability was presented [55]. The organic fertilizer applied to soil might stimulate the decomposition ability of microorganisms. The microorganisms would first decompose macroaggregates. During the decomposition process, not only the extracellular polysaccharides were produced, but also more cementing substances were formed. The binding effect of extracellular polysaccharides through cementing substances would improve the integration of soil minerals and particles, which was conducive to the cementation of microaggregates and silt and clay fractions into macroaggregates [55].

The 1620/2920 rate indicates the level of SOC decomposition, and the SOC content within recalcitrant components was higher with the larger 1620/2920 rate. In the current research, the significant decreased amounts of alcohol and phenol compounds, and the increased amounts of aliphatic compounds, and the increased 1620/2920 rate in M(c)POM fraction inside macroaggregates under manure addition implied that the aggregation of microaggregates into macroaggregates was greater than the decomposition of macroaggregates, leading to more complicated stabilization in Mollisols. Among them, M(c)POM fraction was the most sensitive indicator and played a decisive role. Similarly, all fractions in the microaggregates exhibited the key role in the process of aggregates turnover, due to the changes of alcohol phenol and aliphatic compound amounts, and the 1620/2920 rate in both Fm-POM and Fm-MOM fractions.

The significant and negative correlation was found between OC content of macroaggregates and 1620/2920 rate, but other fractions had the opposite relationship. The major reason might be that the main components of OC in macroaggregates were polysaccharides and carbohydrates with higher active, while the OC of microaggregates and silt + clay components mostly existed in the form of resistant humic substances with lower activity.

The significant correlation with aliphatic and aromatic compounds within macroaggregates and silt and clay fractions in the current research implies that aliphatic compound amounts were high with more active and fast conversion rates, resulting in a larger labile structure ratio in macroaggregates, while the aromatic compound amounts in microaggregates was higher with weaker activity, thus the stability was strong, but the conversion rate was slower. This finding was in line with the conclusions of Steffens et al. on the soil aggregates hierarchy theory [68]. They verified that the amount of aromatic C enhanced and accompanied by the smaller aggregate particle sizes, and the aromatic C with strong resistance to decomposition would selectively gather smaller particle size components in the soil. The present study demonstrates that double-rate manure had no significant impact on maize yield than single-rate manure addition.

According to our findings, a relational graph was proposed showing how manure addition affects aggregate stability in Mollisols (Figure 9). The incremental amounts of aromatic compounds of POM fraction within macroaggregates [M(c)POM] and free microaggregates (Fm-POM) directly increased the MWD value, resulting in enhancing crop yield. Thus, we speculated that physical protection by the occlusion within aggregates of POM could be the critical mechanism for improving aggregate stability after manure addition in Mollisols.

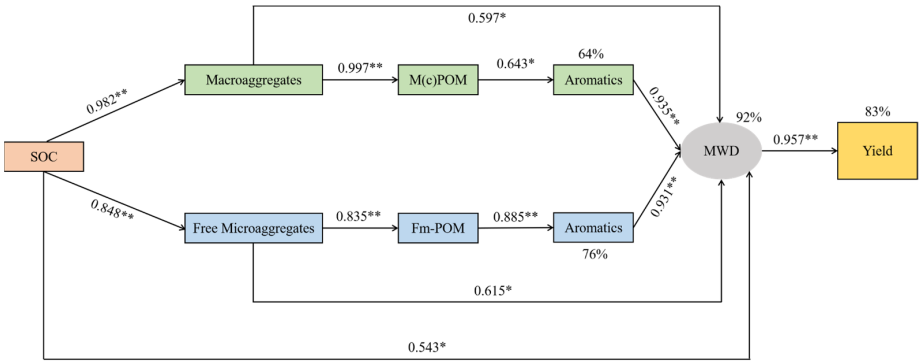


Figure 9. Structural equation model by using OC contents of macroaggregates, free microaggregates, M(c)POM, and Fm-MOM fractions, plus with the amounts of aromatic substances as predictors of soil aggregate stability under 7-year manure addition (p = 0.114, χ^2/df = 1.63, and GFI = 0.927). The numbers next to the arrows indicate the standardized path coefficients, and the solid arrows represent the positive paths, * and ** indicates significant level at $p < 0.05$ and $p < 0.01$, respectively. The percentage of variance for each variable presents next to the box for the corresponding variable. The specific meanings of organic matter and organic carbon fractions with abbreviations are presented in Table 1.

4.4. Selection of Appropriate Fertilization Management

Though significant differences were observed between manure addition and without manure application treatments, the single-rate and double-rate manure addition exhibited no remarkable influences for maize yield components. Additionally, non-fertilizer and only chemical fertilizer treatments presented no significant difference for the pH value, bulk density, SOC, total N, NO_3^--N, Olsen-P, MBC, and MBN in Mollisols between, before, and after crop harvest (Supplementary Table S5). However, the two treatments of manure addition demonstrated the distinct differences for the above-mentioned physical and chemical properties of topsoil between, before, and after crop harvest. Futhermore, some soil indicators including bulk density, NH_4^+-N, SOC, labile OC fractions, aggregate-associated OC contents, and aggregate stability revealed no obvious effect between single-rate and double-rate manure addition. Through integrating previous factors and the economic benefits, the single-rate manure addition of 15 t ha^{-1} was the effective fertilization management under soybean-maize rotation in Mollisols.

5. Conclusions

Compared with the application of chemical fertilizer alone, 7-year manure addition significantly enhanced SOC content at 0–10 cm depth, which was evidenced from the contribution of increased aromatic compounds. Meanwhile, the labile OC (DOC and EOC) contents was obviously raised at a 0–30 cm soil layer via seven years of manure addition. Furthermore, manure addition improved soil aggregate stability, thus increasing crop yield, which turned out to be the increments in MWD and GMD values at 0–30 cm depth. What was noteworthy was that 7-year manure addition increased aromatic compound amounts of particulate organic matter (POM) within macroaggregates and free microaggregates. Together, physical protection by the occlusion within aggregates of POM was the critical mechanism for aggregate stability under manure addition in Mollisols. The single-rate manure addition (15 t ha^{-1}) was the effective fertilization method to enhance SOC stabilization and crop yield in Mollisols by increasing the aromatic compounds amount of POM fraction within aggregates.

Supplementary Materials: The following are available online at https://www.mdpi.com/article/10.3390/agriculture13010088/s1, Figure S1: Organic carbon contents of hierarchical aggregates in Mollisols associated with different fertilization treatments, Figure S2: Size distribution of soil hierarchical aggregates under different fertilization practices, Figure S3: Fertilization effects on FTIR of bulk Mollisols, Figure S4: Fertilization effects on FTIR of organic matter fractions with aggregate grouping in Mollisols, Figure S5: Fertilization effects on FTIR of organic matter components within macroaggregates of physical-chemical combined grouping in Mollisols, Figure S6: Fertilization effects on FTIR of organic matter fractions within free microaggregates of physical-chemical combined grouping in Mollisols, Table S1: Fertilization effects on relative peak area (%) of characteristic peaks observed in the FTIR analysis of bulk Mollisols, Table S2: Fertilization effects on relative peak area (%) of characteristic peaks in organic matter fractions with aggregate grouping in FTIR analysis of Mollisols, Table S3: Fertilization effects on relative peak area (%) of characteristic peaks in organic matter fractions within macroaggregates in physical-chemical combined grouping in FTIR analysis of Mollisols, Table S4: Fertilization effects on relative peak area (%) of characteristic peaks in organic matter fractions within free microaggregates in physical-chemical combined grouping in FTIR analysis of Mollisols, Table S5: The physical and chemical properties of topsoil (0–20 cm) under different fertilization measures before and after crop harvest.

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