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Aggregate Stability under Long-Term Fertilization Practices: The Case of Eroded Ultisols of South-Central China

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Abstract: Soil aggregate stability is an important aspect of soil function and health. Fertilization could potentially alter soil properties and thereby affect aggregate stability. To determine which fertilizer is useful for improving soil fertility and stabilizing soil aggregates and thereby reducing soil erodibility, we examined three types of fertilizer, and measured how soil organic carbon, carbohydrates, and related soil properties influenced aggregate stability in eroded Ultisols. Treatments included control (CK), mineral fertilizer nitrogen (N), phosphorus (P), potassium (K) (NPK), fertilizer NPK plus straw (NPKS), and farmyard manure (FYM). Aggregate stability was tested according to Le Bissonnais method, involving three disruptive tests: fast wetting (FW), slow wetting (SW), and mechanical breakdown (WS). Total organic carbon, particulate organic carbon, mineral-associated carbon, and cold-water-soluble carbohydrate, hot-water-soluble carbohydrate, and dilute acid hydrolysable carbohydrate were measured, as well as soil intrinsic properties (including pH, bulk density, iron and aluminum oxides). The 12-year fertilization had a larger effect on aggregate stability and related soil properties in a 0–15 cm soil layer, whereas no effect was evident at a soil depth of 15–40 cm. MWD (mean weight diameter) under the three tests decreased with increasing soil depth. Fertilization, especially farmyard manure evidently improved MWD_{FW} and MWD_{WS} at a depth of 0–15 cm. Slaking was the main mechanism of aggregate breakdown in Ultisols studied, followed by mechanical breakdown. Correlation analysis showed that MWD_{FW} and MWD_{WS} at a depth of 0–15 cm increased with the increase of particulate organic carbon, total organic carbon, hot-water-soluble carbohydrate and pH. Furthermore, their interaction with amorphous iron oxides enhanced aggregate stability against slaking or, with amorphous aluminum oxides, modified aggregate stability against mechanical breakdown. Consequently, particulate organic carbon was the dominant cementing agent for aggregation in Ultisols studied, and its combination with pH, amorphous aluminum oxides, amorphous iron oxides, and free aluminum oxides play a synergetic role in stabilizing soil aggregate. Accordingly, farmyard manure or fertilizer NPK plus straw improved soil fertility and the ability to resist slaking.

Keywords: Le Bissonnais' method; mean weight diameter; soil organic carbon; carbohydrate; fertilizer

1. Introduction

Ultisols (locally known as red soil) cover approximately 1.14 million km^2 in tropical and subtropical regions of South-Eastern China, representing the dominant soil in South America and Southeastern Asia [1,2]. Inappropriate soil management practices with intensive land development and utilization, as well as unfavorable soil properties, increase the risk of erosion and have been linked with low productivity, resulting in a great hindrance to the local socioeconomic development [3–5]. Fortunately, the adverse effect of soil erosion and degradation could be offset by improved management practices such as fertilizer input and agronomic management, which potentially could increase soil organic matter (SOM) or soil organic carbon (SOC) and restore soil physical properties, including soil aggregate stability [6–10].

Aggregate stability is considered an important indicator of soil physical quality, affecting soil functions such as soil aeration, the movement and storage of soil water, soil erodibility, and carbon sequestration [11–15]. The primary mechanisms of aggregate breakdown by water are slaking, breakdown by differential swelling, mechanical breakdown by raindrop impact, and physiochemical dispersion [16]. Within a site, the relative importance of these mechanisms mostly depends on the physical and chemical properties of soil, which may be affected by agricultural practices. Most often, improvements in aggregate stability were associated with an increase in soil organic carbon content [17–19]. Many studies have shown that organic amendments (e.g., manure and crop residues) to soil over time not only could maintain or increase soil fertility and SOC accumulation but also improve aggregation [6,14,20–23]. Other workers, however, reported that SOC content and soil structure declined following the continuous application of mineral fertilizer alone [24]. Meanwhile, several studies indicated that aggregate stability did not increase following organic amendments (FYM, vermicompost, and lantana compost) [25].

In fact, alteration of SOC is very slow in agricultural soil, and SOC fractions are likely more sensitive and respond more quickly to soil management practices [26,27]. Increasing evidence has indicated that different SOC fractions perform different functions, participating in the formation and stabilization of soil aggregates in different ways [28–31]. More precisely, SOC fractions extracted by water or acid can better represent soil organic binding components, which are directly involved in the formation and stabilization of aggregates [32]. For example, Bouajila and Gallali (2010) [33] suggested that aggregate stability was more associated with particulate organic carbon (POC) compared to SOC. Roberson et al. (1995) [34] reported that aggregate stability was significantly correlated with soil heavy fraction carbohydrate content. In contrast, weaker correlations have been described between water- or acid-extractable C fractions and aggregate stability. Apart from SOC and its fractions, other soil characteristics such as sesquioxides, texture, and exchangeable cations are involved in soil aggregation and stabilization [19–21,35,36]. For instance, macro-aggregates in red soil tended to increase with the accumulation of SOC and Fe-oxides following the application of manure plus nitrogen fertilizer [37]. Nevertheless, the differences in the quality and quantity of these binding agents after the application of different fertilization over time could effectively indicate soil disaggregation or aggregation.

In China, to meet the food demands of an increasing population, more mineral fertilizers are being applied to the soils since the 1980s, but farmyard manure and straw as sources of plant nutrients have been renewed in recent years due to environmental problems related to the application of mineral fertilizers alone. In the past few years, several studies have previously assessed the influence of different fertilizer application on aggregate stability, SOC and its fractions, and other properties across the world. However, it is impossible to transfer these findings to South-Central China, due to differences in soil type, vegetation type, and climate conditions. In South-Central China, Peng et al. (2015) [19] reported that the contributions of sesquioxides and soil organic matter to aggregation in an Ultisol following long-term fertilization, whereas limited information exists regarding aggregate stability under a different breakdown mechanism and related soil properties following long-term fertilization, particularly in eroded dryland.

Here, we hypothesized that the application of mineral fertilizers and organic materials would change SOC and its fractions, and improve aggregate stability. Specifically, we sought: (i) To quantify the characteristics of SOC fractions, carbohydrate fractions, and aggregate stability (MWD) under a long-term application of fertilization; (ii) to understand the role of SOC fractions, carbohydrate fractions, and other soil properties in aggregate stability. The expected results could identify the key factors influencing aggregate stability and sustainable best fertilizer management practice for agricultural systems in South-Central China.

2. Materials and Methods

2.1. Study Site

The field experiment was initiated at the Red Soil Experimental Station of Huazhong Agricultural University (30°02′ N, 114°21′ E; elevation: 41 m) in Xianning County, Hubei province, China, see Figure 1, in 1998, to improve soil quality and identify the best management practice for sustainable crop production. The region is under a typical subtropical monsoon climate with a rainy season from May to early-July and a dry one from late-July to early-September. The annual mean temperature is around 16.5 °C, annual rainfall averages about 1370 mm, and the annual potential evapotranspiration is 1490 mm. The study site is representative of the regional features of natural resources and land use in South-Central China. The soil is classified as Ultisols developed from Quaternary red clay. Water erosion, especially interrill erosion is the domain form of soil erosion in the study area.

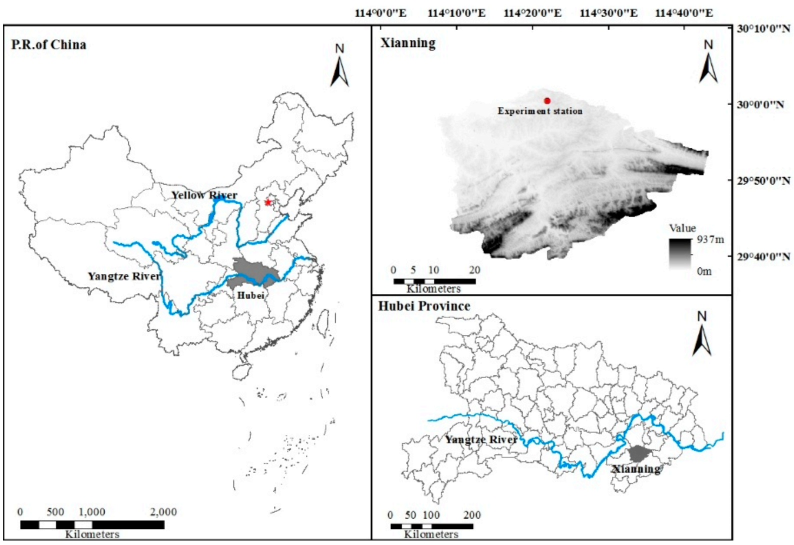

Figure 1. Location of the Red Soil Experimental Station of Huazhong Agricultural University.

2.2. Treatment Details and Management Practices

This long-term field trial originally consisted of four treatments, having a plot size of 3 m × 7 m. The plots were arranged in a randomized block design with three replications. Four fertilizer treatments were included as follows: (1) CK, unfertilized control, (2) NPK, mineral fertilizer (N: 175 kg ha^{-1}, P_2O_5: 150 kg ha^{-1}, K_2O, 115 kg ha^{-1}), (3) NPKS, mineral fertilizer with straw (NPK+1666 kg ha^{-1} straw), and (4) FYM, farmyard manure (10,000 kg ha^{-1} pig or chicken manure, organic matter content 30–35%). The mineral fertilizers were applied in the form of urea, calcium superphosphate, and potassium chloride. Continuous winter wheat (*Triticum aestivum* L.)–summer maize (*Zea mays* L.) rotation was initiated with all phases represented between 1998 and 2011. Fertilizers were applied just before sowing. Throughout the long-term experimentation, all plant species grew rain-fed without any supplementary irrigation. Prior to the experiment, the experimental site was an uncultivated land with low soil quality, which has a thin A horizon resulting from soil erosion.

2.3. Field Sample Collection and Preparation

Soil samples were collected at depths of 0–5 cm, 5–15 cm, 15–25 cm, and 25–40 cm from each plot. The sampled soil was stored in rectangular sampling boxes. Then, soils were air-dried (25–30 °C), crushed, and soil aggregates (>5 mm) were obtained by dry-sieving. Soil pH was measured in a

1:2.5 (soil/water ratio) using a glass electrode. Soil bulk density (BD) was estimated using 100 mL cylinders per depth for each plot. The total porosity (TP) is calculated using the formula, P = 1 − (soil bulk density/soil density), and assuming that the soil density is 2.65 g cm^{-3}. Free (Fe_d and Al_d) and amorphous (Fe_o and Al_o) were determined using citrate-bicarbonate-dithionite (CBD) [38] and ammonium oxalate [39], respectively.

2.4. Extraction of MOC, POC, CH_{HW}, CH_{CW}, and CH_{DA}

Soil organic carbon was determined using dichromate oxidation [40]. The SOC was assumed to be equal to the total C with negligible inorganic C concentration as the soil pH was below 7 [41]. The particulate organic carbon (POC) and mineral-associated organic carbon (MOC) was determined by dispersing with sodium hexametaphosphate using the Cambardella and Elliot's method [42]. The dispersed fraction was sieved through a 53-µm sieve. The retained material was dried and analyzed for organic carbon. MOC is determined as the difference between SOC and POC.

The carbohydrate fractions were determined in the three types of soil extracts, viz. cold-water-soluble (CH_{CW}), hot-water-soluble (CH_{HW}), and dilute-acid-soluble (CH_{DA}) by mixing 10 mL cold distilled water (at 25 °C and shaking for 16 h, CW), hot distilled water (at 85 °C and heated for 2.5 h, HW), and H_2SO_4 (0.25 M and shaking for 16 h, DA). All three types of soil suspension were centrifuged at 3000 rpm for 30 min at 25 °C. Carbohydrate contents of the extract were determined by spectrometry using the phenol-sulfuric acid method at 490 nm with glucose as the standard [43].

2.5. Soil Aggregate Stability Tests

Aggregate stability was determined by the method of Le Bissonnais [16]. The method combines three disruptive tests that correspond to different wetting conditions and energy: fast wetting (FW), slow wetting (SW), and mechanical breakdown (WS). Before experiments, air-dried soil aggregates were dried at 40 °C for 24 h to obtain constant matric potential. For the FW test, the aggregates were immersed in deionized water for 10 min; for the SW test, a similar amount of aggregates were placed on a filter paper and subjected to a tension of −0.3 kPa for 30 min; for the WS test, the aggregates were first immersed in ethanol for 10 min and then transferred into a flask filled with 200 cm^3 of deionized water, subsequently, the flask was corked and agitated end over end 20 times for 1 min. After each test, the fragments for three tests were transferred to a 0.05-mm sieve in ethanol. The slaking resistant aggregates on a 0.05-mm sieve were collected, dried in an oven at 40 °C, and its size distribution was measured by dry sieving with sieves of 5, 2, 1, 0.5, 0.25, 0.1, and 0.05 mm. Three replicates were examined for each test.

The mean weight diameter (MWD) for each treatment was calculated as:

$$\text{MWD} = \sum_{i=1}^{n} d_i w_i \quad (1)$$

where d_i is the mean diameter of the ith sieve size and w_i is the proportion of the total aggregates in the ith fraction. The average of these three indexes was also calculated (MWD_{mean}). The relative slaking index (RSI) and relative mechanical breakdown index (RMI) are used to determine the resistance of slaking and mechanical breakdown of the soil [44].

$$\text{RSI} = \frac{\text{MWD}_{\text{SW}} - \text{MWD}_{\text{FW}}}{\text{MWD}_{\text{SW}}} \times 100 \quad (2)$$

$$\text{RMI} = \frac{\text{MWD}_{\text{SW}} - \text{MWD}_{\text{WS}}}{\text{MWD}_{\text{SW}}} \times 100 \quad (3)$$

2.6. Statistical Analysis

One-way analysis of variance (ANOVA) following least significant difference (LSD) was conducted to determine the statistical significance of fertilization on soil pH, BD, and TP at each soil depth, and multi-way analysis of variance (ANOVA) was carried out to test the effects of fertilization, soil depth, and their interaction on SOC and carbohydrate fractions and aggregate stability indices. Pearson's correlations were performed to explore the relationship between aggregate stability indices and soil physicochemical properties.

Correlation analysis showed that there existed multicollinearity between explanatory variables. To determine the variables that best account for the majority of aggregate stability, stepwise multiple linear regression (MLR) and partial least square regression (PLSR) were used and compared. Four dependent variables (MWD_{FW}, MWD_{WS}, RSI, and RMI) and thirteen independent variables (TOC, POC, MOC, CH_{HW}, CH_{CW}, CH_{DA}, pH, BD, TP, Fe_o, Al_o, Fe_d, and Al_d) were selected. The first three important explanatory variables were selected to predict the response variable according to the Variable Importance Plot (VIP > 1) in PLSR.

One-way and multi-way ANOVA, and stepwise multiple linear regression were performed using the SPSS 18.0 software (SPSS Inc., Chicago, IL, USA). Partial least square regression analysis was carried out using the SIMAC-P 11.5 (Umetric AB, Umea, Sweden), and the remaining figures were made using R (version 3.2.1) software.

3. Results

3.1. Soil pH, Bulk Density, and Total Porosity

Soil was acidic and decreased gradually with soil depth (Table 1). Compared with CK treatment, fertilizer applications (NPK, NPKS, and FYM) raised significantly soil pH at 0–15 cm soil depth. NPKS treatment showed significantly ($P < 0.05$) lower bulk density and significantly ($P < 0.05$) higher total porosity than CK at the 0–15 cm depth. Only slight effects of NPK and FYM treatments on bulk density and total porosity were observed. Irrespective of fertilizers application, bulk density slightly increased with soil depth but total porosity had the inverse trend. Overall, the results indicated that fertilizers application increased soil pH and TP but decreased BD in the 0–15 cm soil layer. No evident differences for pH, BD and TP at 25–40 cm depth for all treatments, however.

Table 1. Soil pH, bulk density, and total porosity under different fertilization management practices.

Soil Properties	Soil Depth (cm)	CK	NPK	NPKS	FYM
pH (1:2.5)	0–5	5.29 d	5.93 c	6.21 a	6.11 b
	5–15	5.36 c	5.53 b	6.16 a	5.61 b
	15–25	5.27 ab	5.35 a	5.21 b	5.08 c
	25–40	5.08 a	5.13 a	5.06 a	4.61 b
Bulk density (g cm^{-3})	0–5	1.20 a	1.18 a	1.15 b	1.19 a
	5–15	1.43 a	1.36 b	1.35 b	1.39 ab
	15–25	1.57 a	1.57 a	1.55 a	1.57 a
	25–40	1.59 a	1.57 a	1.59 a	1.61 a
Total porosity (%)	0–5	54.72 b	55.47 b	56.60 a	55.09 b
	5–15	46.04 b	48.81 a	49.06 a	47.75 ab
	15–25	40.75 a	40.75 a	41.51 a	40.75 a
	25–40	40.00 a	40.75 a	40.00 a	39.25 a

Different letters indicate significance at $P < 0.05$ between different treatments in the same soil layer; CK: control; NPK: mineral fertilizer NPK; NPKS: NPK plus straw; FYM: farmyard manure.

3.2. Soil Organic Carbon and Carbohydrate

The amount of TOC, POC, and MOC was significantly different among soil depths ($P < 0.001$), as shown in Table 2. Fertilizer applications significantly increased the amount of TOC, POC, and MOC compared to the CK when only a depth of 0–5 cm was considered, see Figure 2. However, no significant difference was observed for POC (P = 0.056) or MOC (P = 0.085) when all soil depths were considered, see Table 2; only TOC content showed a significant difference (P = 0.029), see Table 2. Furthermore, fertilizers and soil depths had significant interactive effects on the amount of TOC (P < 0.001) and POC (P < 0.001).

Table 2. Analysis of variance (ANOVA) results of the effects of fertilizers and soil depths on soil organic carbon.

Factor	Source	d.f.	F	Significance
TOC	Fertilizer	3	4.8	0.029
	Soil depth	3	45.5	<0.001
	Fertilizer × Soil depth	9	13.6	<0.001
POC	Fertilizer	3	3.7	0.056
	Soil depth	3	21.4	<0.001
	Fertilizer × Soil depth	9	25.7	<0.001
MOC	Fertilizer	3	3.0	0.085
	Soil depth	3	53.8	<0.001
	Fertilizer × Soil depth	9	1.9	0.083

d.f.: degrees of freedom; F: the ratio of the mean squares between groups and within groups; TOC: total organic carbon; POC: particulate organic carbon; MOC: mineral-associated organic carbon.

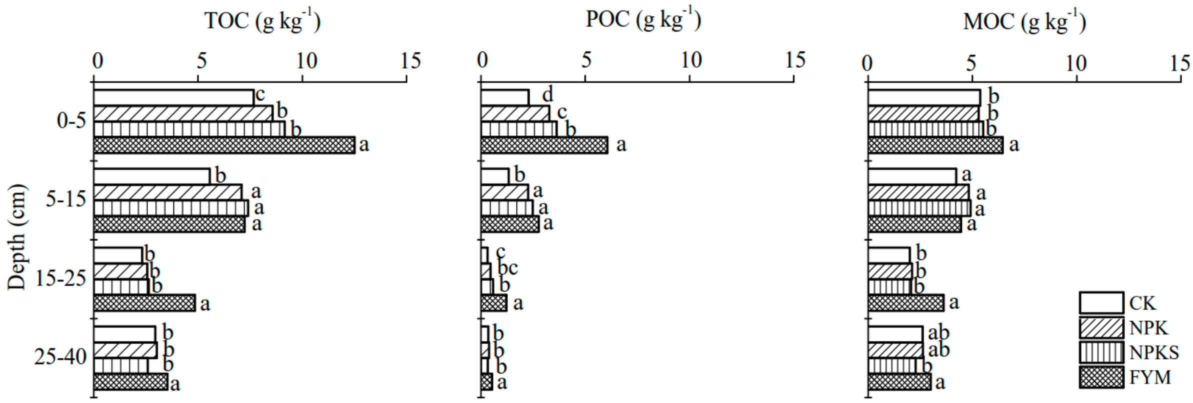

Figure 2. Effects of fertilizer system on soil organic carbon content.

Significant accumulation of TOC, POC, and MOC at a depth of 0–15 cm occurred in accordance with their concentration, see Figure 2 and Table 2. On average, the mean TOC, POC, and MOC contents in the 0–40 cm layer were 14.8–51.9%, 48.9–146.9%, and 3.9–23.3% higher, respectively, after long-term mineral or organic fertilization compared to those in the CK soil. MOC accounted for about 51.5–88.2% of TOC at a depth of 0–40 cm and, therefore, represented the majority of the soil organic carbon, while POC has a minor TOC proportion 11.8–48.5%.

Fertilization does have an effect on the amount of carbohydrate, see Figure 3, although no significant difference was observed among fertilizer treatments at a depth of 0–40 cm, see Table 3. Generally, statistically significant differences (P < 0.05) were obtained with regard to the amount of carbohydrate in each soil layer among fertilizer treatments, especially at a depth of 0–5 cm, see Figure 3. Briefly, the carbohydrate concentration for DA and HW tests at a depth of 0–5 cm ranked in the order: FYM > NPKS > NPK > CK, but for CW test, NPKS resulted in higher levels of carbohydrate.

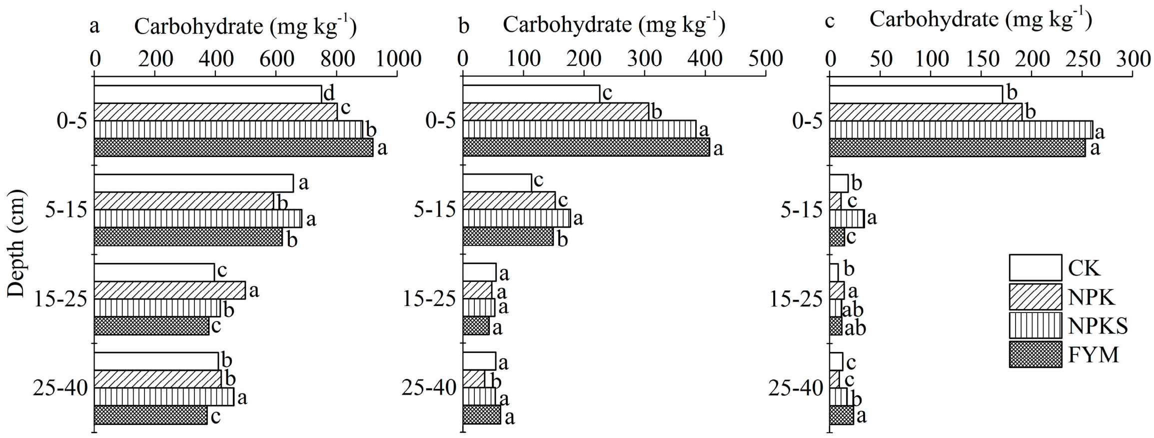

Figure 3. Effects of fertilizer system on the amount of acid-soluble carbohydrate (**a**), hot-water-soluble carbohydrate (**b**), and cold-water-soluble carbohydrate (**c**).

Table 3. ANOVA results of the effects of fertilizers and soil depths on soil carbohydrate.

Factor	Source	d.f.	F	Significance
CH_{HW}	Fertilizer	3	1.3	0.341
	Soil depth	3	37.8	<0.001
	Fertilizer × Soil depth	9	60.77	<0.001
CH_{CW}	Fertilizer	3	1.7	0.238
	Soil depth	3	107.4	<0.001
	Fertilizer × Soil depth	9	10.2	<0.001
CH_{DA}	Fertilizer	3	1.6	0.254
	Soil depth	3	20.5	<0.001
	Fertilizer × Soil depth	9	35.1	<0.001

Notably, the greatest values were found for the acid-soluble carbohydrate concentration (356.5–943.5 mg kg^{-1}), see Figure 3a. The values of the hot water-soluble carbohydrates were lower than those of the acid-soluble carbohydrate at the same soil layer in each plot, see Figure 3b. More obviously, the fraction with the lowest values was the cold water-soluble carbohydrate concentration (8.5–253.2 mg kg^{-1}), see Figure 3c. A significant accumulation of hot-water-soluble carbohydrates at a depth of 0–15 cm occurred, see Figure 3b, whereas cold-water-soluble carbohydrates at a depth of 0–5 cm accumulated in accordance with their concentration, see Figure 3c.

3.3. Aggregate Stability Measured by the Le Bissonnais Method

In general, MWD values decreased with depth in all plots irrespective of the treatment applied, see Table 4 and Figure 5. Considering all MWD related to the three tests (FW, SW, and WS), the aggregate stability for four treatments appeared in general in the order of: MWD_{SW} (slow wetting) > MWD_{WS} (mechanical breakdown) > MWD_{FW} (fast wetting) in each plot, see Figures 4 and 5, indicating that slaking was likely the main mechanism of aggregate breakdown in the soil studied, followed by mechanical breakdown.

Table 4. ANOVA results of the effects of fertilizers and soil depths on aggregate stability.

Treatment	Source	d.f.	F	Significance
FW	Fertilizer	3	2.0	0.182
	Soil depth	3	17.1	<0.001
	Fertilizer × Soil depth	9	341.9	<0.001
SW	Fertilizer	3	4.4	0.037
	Soil depth	3	22.2	<0.001
	Fertilizer × Soil depth	9	40.7	<0.001
WS	Fertilizer	3	6.5	0.012
	Soil depth	3	18.2	<0.001
	Fertilizer × Soil depth	9	37.3	<0.001

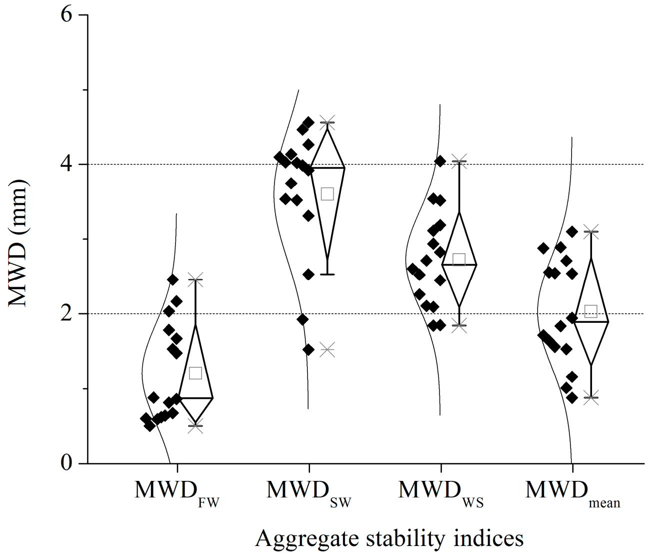

Figure 4. Distribution of mean weight diameter (MWD) under fast wetting (FW), slow wetting (SW), and mechanical breakdown (WS) tests. The diamond boxes indicate the 25th and 75th percentiles; the line in the box indicates the median (50th percentile); "×" indicates outlier values; "□" indicates the average value.

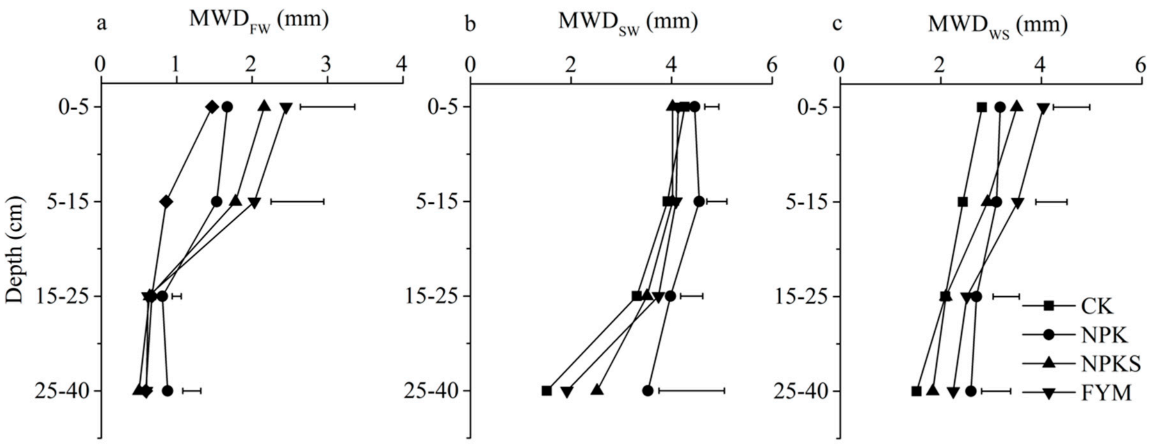

Figure 5. Mean weight diameter (MWD) under different fertilizer treatments with different tests: (**a**) FW, (**b**) SW, and (**c**) WS.

The values of MWD_{SW} were higher with the coefficient of variation of 25%, varying from 1.52 to 4.46 mm. MWD_{WS} ranged from 1.84 to 4.04 mm with a CV of 24%. Relative to MWD_{SW} and MWD_{WS}, MWD_{FW} had relatively lower values (0.50–2.45 mm) but a higher variation of 54% regarding CV, see Figure 4. The MWD_{mean} provided an overall view of aggregate stability at different conditions of soil wetness, ranging from 1.32 to 3.54 mm with an average of 2.51 ± 0.66 mm, see Figure 4. In the 0–15 cm soil layer, MWD_{WS} ranged from 2.46 to 4.04 mm, which corresponded to very stable soil aggregates [17], while MWD_{SW} varied from 3.92 to 4.46 mm, and MWD_{FW} ranged from 0.86 to 2.46 mm. In the 15–40 cm soil layer, MWD_{FW} ranged from 0.50 to 0.88 mm, MWD_{WS} ranged from 1.84 to 2.71 mm, and MWD_{SW} ranged from 1.52 to 3.98 mm, see Figure 5.

After 12 years of fertilizer management, NPK, NPKS, and FYM treatments had significantly ($P < 0.05$) higher MWD_{WS} and MWD_{SW} than CK, see Table 4. As such, NPK, NPKS, and FYM treatments had evidently higher values of MWD_{FW}; although, no significant difference was found ($P = 0.182$). For FW and WS tests, at a depth of 0–5 cm, the FYM plot had the highest MWD, followed by the NPKS plot and NPK plot, and the CK plot had the lowest MWD. Interestingly, the NPK plot has the highest MWD in the same layer for the SW test, see Figure 5b.

3.4. Identification of the Key Factors Influencing Aggregate Stability

As mentioned above, the measured soil properties were affected by fertilization in just the 0–15 cm soil layer, whereas they remained almost unchanged in the 15–40 cm layers. Thus, data collected at the 0–15 cm depth, were used to determine the relationship with aggregate stability indices.

Correlation analysis showed that MWD_{FW} was significantly correlated with TOC ($P = 0.011$), POC ($P = 0.006$), MOC ($P = 0.048$), CH_{HW} ($P = 0.042$), and pH ($P = 0.040$), see Figure 6. Also, MWD_{WS} had significant positive correlations with pH ($P = 0.002$), TOC ($P = 0.007$), POC ($P = 0.002$), and CH_{HW} ($P = 0.048$). However, no apparent relationships were observed between MWD_{SW} and the measured soil properties. MWD_{mean} were significantly and positively correlated with TOC ($P = 0.009$), POC ($P = 0.005$), MOC ($P = 0.044$), and CH_{HW} ($P = 0.048$).

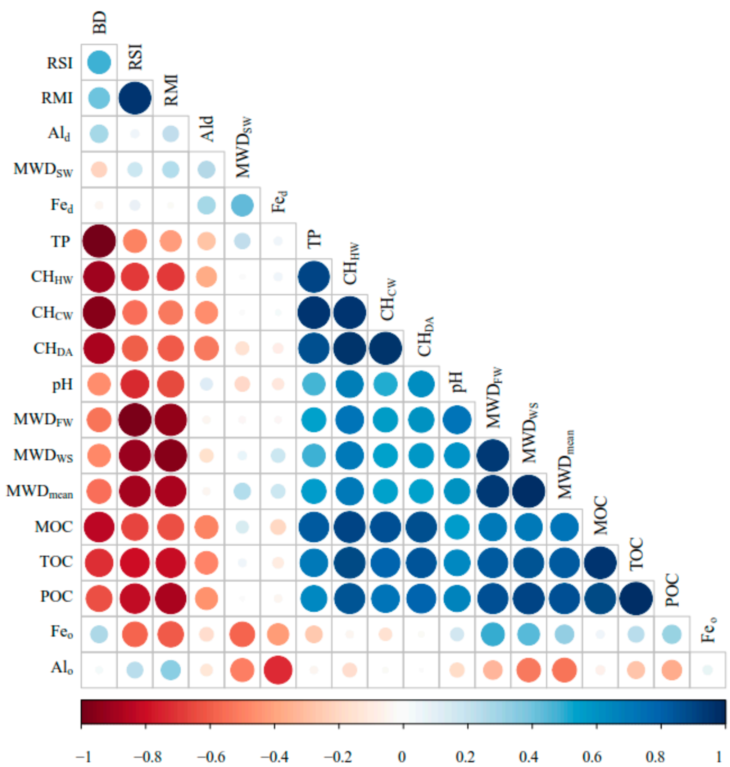

Figure 6. Correlations between aggregate stability and the measured soil properties. The red color means negative correlation while the blue color means positive correlation; the light color and smaller circle mean the lower value of correlation coefficient and the darker color and bigger circle mean the higher value.

Significant correlations also generally existed between TOC, POC, MOC, CH_{HW}, CH_{CW}, and CH_{DA}, see Figure 6. TOC was positively related to TP (P = 0.045) but was negatively related to BD (P = 0.045). Similarly, POC was negatively related to BD but not significantly (P = 0.093). Interestingly, MOC was significantly positive with TP (P = 0.010) and negative with BD (P = 0.010). Notably, no significant relationship (data not shown) was observed between MWD and the soil properties mentioned above in the 15–40 cm layer.

The variables selected for stepwise multiple linear regression (MLR) and partial least square regression (PLSR) analysis are presented in Table 5. The variables included when using MLR and PLSR for each model were a bit different. For MLR, POC together with Al_d contributed to MWD_{FW} significantly (P < 0.01); POC had positive effects on MWD_{WS}; the relative mechanical breakdown index was negatively related to POC; the relative slaking index was also negatively related to POC. For PLSR, POC, Fe_o, and pH were positively associated with MWD_{FW}; and POC, in combination with TOC and Al_o affected MWD_{WS}. For RMI, besides POC, CH_{HW} played a negative role but TOC had a positive role; for RSI, including POC and TOC, Fe_o was selected in the model, explaining 64.1% of its variance.

Table 5. Relationship between aggregate stability indices and soil properties obtained by multiple linear regression (MLR) and partial least square regression (PLSR).

Method	Formulas	Adj.R^2	SEE	Significance
MLR	$MWD_{FW} = 0.368POC + 0.897Al_d - 1.374$	0.857	0.184	0.003
	$MWD_{WS} = 0.315POC + 2.247$	0.786	0.228	0.002
	$RSI = -0.071POC + 0.796$	0.631	0.074	0.011
	$RMI = -0.078POC + 0.470$	0.728	0.065	0.004
PLSR	$MWD_{FW} = 0.198POC + 0.411pH + 1.560Fe_o - 2.701$	0.745	0.246	0.038
	$MWD_{WS} = 0.748POC - 0.310TOC - 0.562Al_o + 3.930$	0.801	0.220	0.123
	$RSI = -0.054POC - 0.005TOC - 0.525Fe_o + 1.284$	0.641	0.072	0.073
	$RMI = -0.285POC + 0.157TOC - 0.0002CH_{HW} - 0.121$	0.786	0.058	0.027

SEE: Standard error of the estimate.

4. Discussion

4.1. Influence of Fertilizer on Soil Properties

In this study, different fertilization practices have a larger effect on the surface layer, clearly affecting soil pH, BD, TP, organic carbon fractions, carbohydrate, and MWD at a soil depth of 0–15 cm, see Tables 1–3 and Figures 2–5. In general, the amendments of manure, mineral fertilizer, and mineral fertilizer plus straw had a tendency to deacidify the present soil, see Table 1, particularly in plots under FYM; soil pH was significantly higher than that in CK. Similarly, in some studies [21,45,46], the application of farmyard manure or fertilizer NPK plus straw increased soil pH. However, soil pH has a tendency to decrease following the application of organic manure, mineral fertilizer, and straw in several studies [47]. The contradictory results could be explained by the observation of Xu et al. (2006) [48], in which the extent of the soil pH increase depended on the initial pH and buffer capacity of the soil and the rate of decomposition and association/dissociation of organic compounds in plant residues.

Not surprisingly, NPK and FYM slightly decreased BD and increased TP in comparison with CK, but BD and TP in FYM were significantly different from CK. Similarly, decreased BD and increased TP with inorganic or organic fertilization were presented in other studies [35,49–52]. For NPK plus straw, the improvement of BD and TP can be partially ascribed to the increased SOC because BD was negatively correlated with SOC content. Correspondingly, the incorporation of straw could lead to soil particles to stick together, which in turn, form soil aggregates. Similar results have been reported [14,53–55].

In this study, the TOC contents in all treatments ranged from 5.6 to 12.5 g kg^{-1} at a depth of 0–15 cm after 12 years of intensive cropping. The greatest TOC or POC content was found in FYM treatment, typically at a depth of 0–5 cm, followed by NPKS treatment and then by NPK treatment, see Figure 1. Likewise, various past studies have reported similar results due to the addition of manure, straw, and NPK fertilizers in long-term fertilization experiments elsewhere [27,56–60]. Accordingly, the superior effect of FYM over NPKS and NPK in this study may be ascribed to two aspects: (i) the increased input of organic materials into soil compared with no manure treatments; (ii) the manure-induced soil aggregation would be helpful for slowing down the organic matter turnover rate [61,62]. Also, the accumulation of SOC and POC content at the topsoil in the NPKS plot may be attributed to the greater input amount of organic matter by incorporation and decomposition of straw [63,64]. As with the improvement in SOC and POC under NPK mineral fertilizers alone, in our study, two crops per year maintained more roots and rhizodeposition; although, the aboveground straws were removed whereas there was considerable stubble biomass input. Therefore, NPK mineral fertilizers alone could increase the level of POC by better crop growth relative to CK treatment to a large extent. Similar positive effects of different combinations of N, P, and K on SOC have been previously reported [65]. In all, the improvement may be partly attributed to the direct effects of the manure and indirect effects of straw addition and stubble residues in our study.

Soil carbohydrates, as important parts of soil organic matter, are the main component of the labile pool of SOM. As expected, the performances of the three fertilizer treatments differed in terms of their contribution to carbohydrate concentrations. CH_{DA} and CH_{HW} concentrations at a depth of 0–5 cm decreased in the order: FYM > NPKS > NPK > CK. Somehow, the highest CH_{CW} at a depth of 0–5 cm was recorded under NPKS treatment. In line with our findings, the application of manure, organic materials, and mineral fertilizer generally increased soil carbohydrates [66,67]. The improvement can be partially explained by differences in the amendments of carbohydrates in manure and plant-derived carbohydrates. Moreover, high amounts of carbohydrate in FYM and NPKS seem to be the result of the higher amount of manure and greater crop yield and hence, a higher return of crop residues in comparison with CK [68,69]. Similar to the results obtained by Adesodun et al. (2001) [28], the carbohydrate concentrations ranked in the order, $CH_{DA} > CH_{HW} > CH_{CW}$. As stated above, the general trend in the concentration of all three carbohydrates declined with soil depth. Overall, the effect of the amendments of manure, organic materials, and mineral fertilizer on organic carbon and carbohydrate was lower in the deeper layers relative to the surface layers of the soil.

4.2. Aggregate Stability and Its Relationship with Other Properties

The soil aggregate stability values (MWD) in this study showed relatively large variability, see Figures 4 and 5. Independent analyses using three tests of the Le Bissonnais method can contribute to a better understanding of the mechanisms involved in aggregate stability. MWD values followed the order of $MWD_{SW} > MWD_{WS} > MWD_{FW}$, indicating that the FW test was the most efficient for disrupting soil aggregates. Thus, slaking was the main mechanism of aggregate breakdown in the soil studied, followed by mechanical breakdown, which agrees with other studies in the same area [70].

Improvements in aggregate stability following additional amendments have been already reported [8,71]. As we revealed above, fertilizer amendments obviously affected soil aggregate stability at a depth of 0–15 cm, see Figure 5. In general, aggregate stability was affected by intrinsic properties (e.g., organic carbon or fractions, soil texture, mineralogy, and pH) and external factors (e.g., climate, agricultural management, and vegetation types) [35,72]. In this study, there is no difference regarding vegetation type (maize and wheat) and climate factor; therefore, agricultural management and soil properties were the main factors.

Amendments of manure and straw residues have been previously reported as being the effective method for improving aggregate stability and soil fertility [54,73,74]. As stated above, the long-term application of FYM and NPKS increased the MWD and promoted soil aggregation in the 0–15 cm soil layer, see Figure 5. The positive effects of FYM and NPKS on aggregate stability may be attributed mainly to the improvement of soil organic carbon through a greater amount of organic inputs. Mikha et al. (2010) [75] pointed out that long-term manure application promoted the formation of macro-aggregates and increased aggregate stability. Also, amendments may indirectly affect aggregate stability by increasing crop productivity and biological activity. Furthermore, more root stubble and biomass added to the soil under NPKS and FYM plots exerted a considerable influence on aggregate stability to some degree due to higher levels of binding agents such as polysaccharides and fungi. Likewise, as discussed above, mineral fertilizer alone might help in increasing root biomass compared to CK, which ultimately increased the MWD value [49,76]. In all, such effects with different fertilization treatments on aggregate stability can be ascribed to their impacts through organic matter input on soil properties.

Soil organic carbon or its fractions such as labile carbon and biologically active carbon plays a key role in aggregate stability [63,72]. POC was directly related to soil aggregation since it can form an organic core surrounded by clay, silt particles, and aggregates [11]. In addition, it has been found that carbohydrates were important aggregate-binding agents [77]. Similarly, in our experiment, POC, TOC, and CH_{HW} were significantly correlated with aggregate stability (FW and WS), indicating that POC, TOC, and CH_{HW} contribute to aggregate stabilization of soil particles, see Figure 6. More precisely, MWD was more associated with POC compared to TOC and CH_{HW}, see Figure 6, in line with the finding of Bouajila and Gallali (2010) [33]. Zotarelli et al. (2007) [78] suggested that the stabilization of particulate organic matter within microaggregates was one of the major mechanisms for SOC protection. Additionally, glomalin-related soil proteins (GRSR) could contribute to the formation and maintenance of soil aggregates following fertilizers amendments [79]. Further study on this topic would improve our understanding of the relationships between POC, GRSR, carbohydrate, and aggregate stability following long-term fertilizers practices.

Consistently with our studies, many workers have found that CH_{HW} was more closely related to aggregate stability than CH_{CA} [29,80]. Unlike our studies, CH_{HW} had a closer relationship with aggregate stability compared to SOC in the study of Yousefi et al. (2008) [29]. The discrepancy was, in part, attributed to soil properties variation, indirectly changing the relationship between organic matter fractions and aggregate stability [33]; further, this relationship varied widely between soils. Interestingly, soil pH was strongly correlated with MWD_{FW}, see Figure 6. Moreover, in the partial least square regression (PLSR), pH together with POC and Fe_o, contributed to 74.5% variations of MWD_{FW}, see Table 5, implying that pH, POC, and Fe_o and their interaction may be an important mechanism for aggregate stability against slaking. In agreement with the findings of Regelink et al. (2015) [81], the effect of pH on aggregate stability may be attributed to the enhanced coagulation of organically-coated particles at a relatively lower pH level in this study. Formation of Al-humus complexes mainly occurs at a lower soil pH, which may favor aggregate stability. Multiple stepwise linear regression (MLR) results showed that the combination of POC and Al_d significantly improved aggregate stability against slaking, although the effect of Al_d on soil aggregation was absent, see Table 5 and Figure 6. The potential influence of Al_d on the aggregate stability of Ultisols has been confirmed in another study [82].

Compared with slaking, only POC contributed positively to MWD_{WS} in MLR, while POC, TOC, and Al_o in PLSR, played a positive and negative role in aggregate stability against mechanical breakdown, respectively, accounting together for 80.1% in MWD_{WS}. Wu et al. (2017) [83] found that amorphous aluminum oxides excluding complex forms contributed negatively to MWD_{WS} in MLR. Owing to clay fractions in the Ultisols studied was dominant in kaolinite, the bonding materials between aggregates involved not only organic materials but also Fe and Al oxides and other cementing materials [11]. Furthermore, several researchers reported that soil oxides acted as important binding agents and play important roles in aggregate formation in Ultisols from subtropical China [44,82]. As discussed above, our results meant that Al_d, Fe_o, and Al_o alone might not play key roles in soil aggregation; in association with POC, TOC, or pH, they may be indispensable components in soil aggregation. Furthermore, Amézketa (1999) [8] indicated that Fe and Al oxides, which bond with organic and inorganic compounds or aggregates through a cation bridge to improve soil structure, are the most active components in soil.

As stated above, the importance and contribution of soil organic carbon or fractions of carbon to aggregate formation and stability have been well reported. Plante and McGill (2002) [84] found that macro-aggregation and micro-aggregation could be enhanced by increasing the SOC content. Furthermore, Fe and Al oxides seemed to be major agents of <0.25 mm aggregates in Ultisols, whereas soil organic matter played a primary role in stabilizing lager aggregates (0.25–2.00 mm) [19]. A further indication of the role of SOM comes from the study of Peng et al. (2015) [19], involved in improving the soil aggregation by increasing the carbon input. Here, the improvement of aggregate stability resulted, in part, from directly and indirectly increasing the soil organic carbon content. Furthermore, previous discussions in this and other publications point out that POC had a greater effect on soil aggregation than SOC and other organic carbon fractions, see Table 5 and Figure 6. Apart from the soil physio-chemical properties mentioned above, the relatively larger root biomass and root length density of maize or wheat under different fertilizer treatments compared to CK influenced most aggregate stability [85,86]. Regardless, our results indicated that POC was the dominant cementing agent for aggregation in Ultisols under long-term fertilization regimes; Al_d, Fe_o, Al_o, and pH were also effective in binding aggregates when they interact with POC.

5. Conclusions

Long-term fertilization treatments, especially FYM application, evidently affected soil pH, bulk density, SOC, carbohydrate and aggregate formation, and stability (MWD). Fertilizations had positive and variable effects on TOC, POC, CH_{DA}, CH_{HW}, and CH_{CW} in the 0–15 cm soil layer. A great improvement in the values of MWD indicated fertilization treatments had positive effects on aggregate stability, and with regard to the treatments with all fertilizers and CK, FYM application resulted in further beneficial effects. The acid-soluble soil carbohydrates were greater than other carbohydrate fractions. Slaking is likely the main disruptive force of aggregate breakdown in Ultisols studied.

Aggregate stability against slaking was controlled by POC, pH, and Fe_o, while POC, TOC, and Al_o influenced aggregate stability against mechanical breakdown. Aggregate stability against slaking or mechanical breakdown decreased with soil depth. POC, a strong binding agent, is a better indicator of aggregate stability, and together with other factors such as Fe and Al oxides and pH, contributed to soil aggregation and structure. According to this study, farmyard manure or NPK mineral fertilizer plus straw appears to be the recommended strategy for the Ultisols of the study area, not only improving soil fertility and soil structure but also increasing carbon sequestration and the ability to resist erosion. Future work should focus more on how the interactions between POC, sesquioxides, and pH influence soil aggregation.

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References

1. Zhang, B.; Yang, Y.S.; Zepp, H. Effect of vegetation restoration on soil and water erosion and nutrient losses of a severely eroded clayey Plinthudult in southeastern China. *Catena* **2004**, *57*, 77–90. [CrossRef]
2. Wang, J.G.; Li, Z.X.; Cai, C.F.; Yang, W.; Ma, R.M.; Zhang, G.B. Effects of stability, transport distance and two hydraulic parameters on aggregate abrasion of Ultisols in overland flow. *Soil Tillage Res.* **2013**, *126*, 134–142. [CrossRef]
3. Guo, Z.L.; Ma, M.J.; Cai, C.F.; Wu, Y.W. Combined effects of simulated rainfall and overland flow on sediment and solute transport in hillslope erosion. *J. Soil Sediment* **2018**, *18*, 1120–1132. [CrossRef]

4. Zhang, B.; Zhang, T.L.; Zhao, Q.G. Soil Erosion of Various Farming Systems in Subtropical China. *Pedosphere* **1996**, *6*, 225–233.
5. Nie, X.D.; Li, Z.W.; Huang, J.Q.; Liu, L.; Xiao, H.B.; Liu, C.; Zeng, G.M. Thermal stability of organic carbon in soil aggregates as affected by soil erosion and deposition. *Soil Tillage Res.* **2018**, *175*, 82–90. [CrossRef]
6. Huang, S.; Peng, X.X.; Huang, Q.R.; Zhang, W.J. Soil aggregation and organic carbon fractions affected by long-term fertilization in a red soil of subtropical China. *Geoderma* **2010**, *154*, 364–369. [CrossRef]
7. Tejada, M.; Gonzalez, J.L. Influence of two organic amendments on the soil physical properties, soil losses, sediments and runoff water quality. *Geoderma* **2008**, *145*, 325–334. [CrossRef]
8. Amezketa, E. Soil aggregate stability: A review. *J. Sustain. Agric.* **1999**, *14*, 83–151. [CrossRef]
9. He, Y.T.; He, X.H.; Xu, M.G.; Zhang, W.J.; Yang, X.Y.; Huang, S.M. Long-term fertilization increases soil organic carbon and alters its chemical composition in three wheat-maize cropping sites across central and south China. *Soil Tillage Res.* **2018**, *177*, 79–87. [CrossRef]
10. Zhou, H.; Fang, H.; Mooney, S.J.; Peng, X.H. Effects of long-term inorganic and organic fertilizations on the soil micro and macro structures of rice paddies. *Geoderma* **2016**, *266*, 66–74. [CrossRef]
11. Six, J.; Elliott, E.T.; Paustian, K. Soil structure and soil organic matter: II. A normalized stability index and the effect of mineralogy. *Soil Sci. Soc. Am. J.* **2000**, *64*, 1042–1049. [CrossRef]
12. Shi, P.; Arter, C.; Liu, X.Y.; Keller, M.; Schulin, R. Soil aggregate stability and size-selective sediment transport with surface runoff as affected by organic residue amendment. *Sci. Total Environ.* **2017**, *607*, 95–102. [CrossRef] [PubMed]
13. Toosi, E.R.; Kravchenko, A.N.; Mao, J.; Quigley, M.Y.; Rivers, M.L. Effects of management and pore characteristics on organic matter composition of macroaggregates: Evidence from characterization of organic matter and imaging. *Eur. J. Soil Sci.* **2017**, *68*, 200–211. [CrossRef]
14. Zhang, P.; Wei, T.; Jia, Z.K.; Han, Q.F.; Ren, X.L. Soil aggregate and crop yield changes with different rates of straw incorporation in semiarid areas of northwest China. *Geoderma* **2014**, *230*, 41–49. [CrossRef]
15. Lal, R. Soil degradation by erosion. *Land Degrad. Dev.* **2001**, *12*, 519–539. [CrossRef]
16. Le Bissonnais, Y. Aggregate stability and assessment of soil crustability and erodibility. I. Theory and methodology. *Eur. J. Soil Sci.* **1996**, *47*, 425–437. [CrossRef]
17. Chaney, K.; Swift, R.S. The Influence of Organic-Matter on Aggregate Stability in Some British Soils. *J. Soil Sci.* **1984**, *35*, 223–230. [CrossRef]
18. Haydu-Houdeshell, C.A.; Graham, R.C.; Hendrix, P.F.; Peterson, A.C. Soil aggregate stability under chaparral species in southern California. *Geoderma* **2018**, *310*, 201–208. [CrossRef]
19. Peng, X.; Yan, X.; Zhou, H.; Zhang, Y.Z.; Sun, H. Assessing the contributions of sesquioxides and soil organic matter to aggregation in an Ultisol under long-term fertilization. *Soil Tillage Res.* **2015**, *146*, 89–98. [CrossRef]
20. Zhang, J.G.; Bo, G.D.; Zhang, Z.F.; Kong, F.Y.; Wang, Y.; Shen, G.M. Effects of Straw Incorporation on Soil Nutrients, Enzymes, and Aggregate Stability in Tobacco Fields of China. *Sustainability* **2016**, *8*. [CrossRef]
21. Tripathi, R.; Nayak, A.K.; Bhattacharyya, P.; Shukla, A.K.; Shahid, M.; Raja, R.; Panda, B.B.; Mohanty, S.; Kumar, A.; Thilagam, V.K. Soil aggregation and distribution of carbon and nitrogen in different fractions after 41 years long-term fertilizer experiment in tropical rice-rice system. *Geoderma* **2014**, *213*, 280–286. [CrossRef]
22. Bandyopadhyay, P.K.; Saha, S.; Mani, P.K.; Mandal, B. Effect of organic inputs on aggregate associated organic carbon concentration under long-term rice–wheat cropping system. *Geoderma* **2010**, *154*, 379–386. [CrossRef]
23. Singh, G.; Jalota, S.K.; Singh, Y. Manuring and residue management effects on physical properties of a soil under the rice-wheat system in punjab, india. *Soil Tillage Res.* **2007**, *94*, 229–238. [CrossRef]
24. Fonte, S.J.; Yeboah, E.; Ofori, P.; Quansah, G.W.; Vanlauwe, B.; Six, J. Fertilizer and Residue Quality Effects on Organic Matter Stabilization in Soil Aggregates. *Soil Sci. Soc. Am. J.* **2009**, *73*, 961–966. [CrossRef]
25. Abiven, S.; Menasseri, S.; Angers, D.A.; Leterme, P. Dynamics of aggregate stability and biological binding agents during decomposition of organic materials. *Eur. J. Soil Sci.* **2007**, *58*, 239–247. [CrossRef]
26. Tirol-Padre, A.; Ladha, J.K. Assessing the reliability of permanganate-oxidizable carbon as an index of soil labile carbon. *Soil Sci. Soc. Am. J.* **2004**, *68*, 969–978. [CrossRef]
27. Xu, M.G.; Lou, Y.L.; Sun, X.L.; Wang, W.; Baniyamuddin, M.; Zhao, K. Soil organic carbon active fractions as early indicators for total carbon change under straw incorporation. *Biol. Fert. Soils* **2011**, *47*, 745–752. [CrossRef]

28. Adesodun, J.K.; Mbagwu, J.S.C.; Oti, N. Structural stability and carbohydrate contents of an ultisol under different management systems. *Soil Tillage Res.* **2001**, *60*, 135–142. [CrossRef]
29. Yousefi, M.; Hajabbasi, M.; Shariatmadari, H. Cropping system effects on carbohydrate content and water-stable aggregates in a calcareous soil of Central Iran. *Soil Tillage Res.* **2008**, *101*, 57–61. [CrossRef]
30. Roberson, E.B.; Sarig, S.; Firestone, M.K. Cover Crop Management of Polysaccharide-Mediated Aggregation in an Orchard Soil. *Soil Sci. Soc. Am. J.* **1991**, *55*, 734–739. [CrossRef]
31. Ball, B.C.; Cheshire, M.V.; Robertson, E.A.G.; Hunter, E.A. Carbohydrate composition in relation to structural stability, compactibility and plasticity of two soils in a long-term experiment. *Soil Tillage Res.* **1996**, *39*, 143–160. [CrossRef]
32. Haynes, R.J. Labile organic matter fractions as central components of the quality of agricultural soils: An overview. *Adv. Agron.* **2005**, *85*, 221–268.
33. Bouajila, A.; Gallali, T. Land use effect on soil and particulate organic carbon, and aggregate stability in some soils in Tunisia. *Afr. J. Agric. Res.* **2010**, *5*, 764–774.
34. Roberson, E.B.; Sarig, S.; Shennan, C.; Firestone, M.K. Nutritional Management of Microbial Polysaccharide Production and Aggregation in an Agricultural Soil. *Soil Sci. Soc. Am. J.* **1995**, *59*, 1587–1594. [CrossRef]
35. Bronick, C.J.; Lal, R. Soil structure and management: A review. *Geoderma* **2005**, *124*, 3–22. [CrossRef]
36. Bongiovanni, M.D.; Lobartini, J.C. Particulate organic matter, carbohydrate, humic acid contents in soil macro- and microaggregates as affected by cultivation. *Geoderma* **2006**, *136*, 660–665. [CrossRef]
37. Yin, Y.; Wang, L.; Liang, C.H.; Xi, F.M.; Pei, Z.J.; Du, L.Y. Soil Aggregate Stability and Iron and Aluminium Oxide Contents Under Different Fertiliser Treatments in a Long-Term Solar Greenhouse Experiment. *Pedosphere* **2016**, *26*, 760–767. [CrossRef]
38. Mehra, O.P.; Jackson, M.L. Iron oxides removal from soils and clays. Dithionite-citrate systems buffered with sodium bicarbonate. *Clay Clay Miner.* **1960**, *7*, 317–327. [CrossRef]
39. Mckeague, J.A.; Day, D.H. Dithionite- and Oxalate-Extractable Fe and Al as Aids in Differentiating Various Classes of Soils. *Can. J. Soil Sci.* **1966**, *46*, 13–22. [CrossRef]
40. Nelson, D.W.; Sommers, L.E. Total carbon, organic carbon and organic matter. In *Methods of Soil Analysis*, Part 2, 2nd ed.; American Society of Agronomy: Madison, WI, USA, 1982; pp. 539–577.
41. Jagadamma, S.; Lal, R. Distribution of organic carbon in physical fractions of soils as affected by agricultural management. *Biol. Fert. Soils* **2010**, *46*, 543–554. [CrossRef]
42. Cambardella, C.A.; Elliott, E.T. Particulate Soil Organic-Matter Changes across a Grassland Cultivation Sequence. *Soil Sci. Soc. Am. J.* **1992**, *56*, 777–783. [CrossRef]
43. Dubois, M.; Gilles, K.A.; Hamilton, J.K.; Rebers, P.A.; Smith, F. Colorimetric Method for Determination of Sugars and Related Substances. *Anal. Chem.* **1956**, *28*, 350–356. [CrossRef]
44. Zhang, B.; Horn, R. Mechanisms of aggregate stabilization in Ultisols from subtropical China. *Geoderma* **2001**, *99*, 123–145. [CrossRef]
45. Whalen, J.K.; Chang, C.; Clayton, G.W.; Carefoot, J.P. Cattle manure amendments can increase the pH of acid soils. *Soil Sci. Soc. Am. J.* **2000**, *64*, 962–966. [CrossRef]
46. Qiu, S.J.; Gao, H.J.; Zhu, P.; Hou, Y.P.; Zhao, S.C.; Rong, X.M.; Zhang, Y.P.; He, P.; Christie, P.; Zhou, W. Changes in soil carbon and nitrogen pools in a Mollisol after long-term fallow or application of chemical fertilizers, straw or manures. *Soil Tillage Res.* **2016**, *163*, 255–265. [CrossRef]
47. Gong, W.; Yan, X.Y.; Wang, J.Y.; Hu, T.X.; Gong, Y.B. Long-term manuring and fertilization effects on soil organic carbon pools under a wheat-maize cropping system in North China Plain. *Plant Soil* **2009**, *314*, 67–76. [CrossRef]
48. Xu, J.M.; Tang, C.; Chen, Z.L. The role of plant residues in pH change of acid soils differing in initial pH. *Soil Biol. Biochem.* **2006**, *38*, 709–719. [CrossRef]
49. Rasool, R.; Kukal, S.S.; Hira, G.S. Soil organic carbon and physical properties as affected by long-term application of FYM and inorganic fertilizers in maize-wheat system. *Soil Tillage Res.* **2008**, *101*, 31–36. [CrossRef]
50. Xin, X.L.; Zhang, J.B.; Zhu, A.N.; Zhang, C.Z. Effects of long-term (23 years) mineral fertilizer and compost application on physical properties of fluvo-aquic soil in the North China Plain. *Soil Tillage Res.* **2016**, *156*, 166–172. [CrossRef]

51. Chaudhary, S.; Dheri, G.S.; Brar, B.S. Long-term effects of NPK fertilizers and organic manures on carbon stabilization and management index under rice-wheat cropping system. *Soil Tillage Res.* **2017**, *166*, 59–66. [CrossRef]
52. Liu, S.L.; Huang, D.Y.; Chen, A.L.; Wei, W.X.; Brookes, P.C.; Li, Y.; Wu, J.S. Differential responses of crop yields and soil organic carbon stock to fertilization and rice straw incorporation in three cropping systems in the subtropics. *Agric. Ecosyst. Environ.* **2014**, *184*, 51–58. [CrossRef]
53. Mulumba, L.N.; Lal, R. Mulching effects on selected soil physical properties. *Soil Tillage Res.* **2008**, *98*, 106–111. [CrossRef]
54. Wei, S.F.; Gao, M.; Shao, J.A.; Xie, D.T.; Pan, G.X. Soil aggregate and its response to land management practices. *China Particuol.* **2006**, *4*, 211–219. [CrossRef]
55. Huang, R.; Lan, M.L.; Liu, J.; Gao, M. Soil aggregate and organic carbon distribution at dry land soil and paddy soil: The role of different straws returning. *Environ. Sci. Pollut. Res.* **2017**, *24*, 27942–27952. [CrossRef] [PubMed]
56. Tian, K.; Zhao, Y.C.; Xu, X.H.; Hai, N.; Huang, B.A.; Deng, W.J. Effects of long-term fertilization and residue management on soil organic carbon changes in paddy soils of China: A meta-analysis. *Agric. Ecosyst. Environ.* **2015**, *204*, 40–50. [CrossRef]
57. Dou, X.L.; He, P.; Cheng, X.L.; Zhou, W. Long-term fertilization alters chemically-separated soil organic carbon pools: Based on stable C isotope analyses. *Sci. Rep.-UK* **2016**, *6*. [CrossRef] [PubMed]
58. Peng, X.; Zhu, Q.H.; Xie, Z.B.; Darboux, F.; Holden, N.M. The impact of manure, straw and biochar amendments on aggregation and erosion in a hillslope Ultisol. *Catena* **2016**, *138*, 30–37. [CrossRef]
59. Manna, M.C.; Swarup, A.; Wanjari, R.H.; Mishra, B.; Shahi, D.K. Long-term fertilization, manure and liming effects on soil organic matter and crop yields. *Soil Tillage Res.* **2007**, *94*, 397–409. [CrossRef]
60. Naveed, M.; Moldrup, P.; Vogel, H.J.; Lamande, M.; Wildenschild, D.; Tuller, M.; de Jonge, L.W. Impact of long-term fertilization practice on soil structure evolution. *Geoderma* **2014**, *217*, 181–189. [CrossRef]
61. Marriott, E.E.; Wander, M. Qualitative and quantitative differences in particulate organic matter fractions in organic and conventional farming systems. *Soil Biol. Biochem.* **2006**, *38*, 1527–1536. [CrossRef]
62. Su, Y.Z.; Wang, F.; Suo, D.R.; Zhang, Z.H.; Du, M.W. Long-term effect of fertilizer and manure application on soil-carbon sequestration and soil fertility under the wheat-wheat-maize cropping system in northwest China. *Nutr. Cycl. Agroecosys.* **2006**, *75*, 285–295. [CrossRef]
63. Abiven, S.; Menasseri, S.; Chenu, C. The effects of organic inputs over time on soil aggregate stability—A literature analysis. *Soil Biol. Biochem.* **2009**, *41*, 1–12. [CrossRef]
64. Guo, Z.L.; Zhao, S.H.; Jiang, J.; Cai, C.F. Nitrogen mineralization controlled by N/P ratio of plant residues from riparian buffer strip. *Eur. J. Soil Biol.* **2015**, *67*, 5–11.
65. Yadav, R.L.; Yadav, D.S.; Singh, R.M.; Kumar, A. Long term effects of inorganic fertilizer inputs on crop productivity in a rice-wheat cropping system. *Nutr. Cycl. Agroecosys.* **1998**, *51*, 193–200. [CrossRef]
66. Yin, Y.; Liang, C.H.; Pei, Z.J. Effect of greenhouse soil management on soil aggregation and organic matter in northeast China. *Catena* **2015**, *133*, 412–419. [CrossRef]
67. Brar, B.S.; Singh, K.; Dheri, G.S.; Balwinder, K. Carbon sequestration and soil carbon pools in a rice-wheat cropping system: Effect of long-term use of inorganic fertilizers and organic manure. *Soil Tillage Res.* **2013**, *128*, 30–36. [CrossRef]
68. Xiao, C.; Fauci, M.; Bezdicek, D.F.; McKean, W.T.; Pan, W.L. Soil microbial responses to potassium-based black liquor from straw pulping. *Soil Sci. Soc. Am. J.* **2006**, *70*, 72–77. [CrossRef]
69. Debosz, K.; Petersen, S.O.; Kure, L.K.; Ambus, P. Evaluating effects of sewage sludge and household compost on soil physical, chemical and microbiological properties. *Appl. Soil Ecol.* **2002**, *19*, 237–248. [CrossRef]
70. Ma, R.M.; Cai, C.F.; Li, Z.X.; Wang, J.G.; Xiao, T.Q.; Peng, G.Y.; Yang, W. Evaluation of soil aggregate microstructure and stability under wetting and drying cycles in two Ultisols using synchrotron-based X-ray micro-computed tomography. *Soil Tillage Res.* **2015**, *149*, 1–11. [CrossRef]
71. Caravaca, F.; Lax, A.; Albaladejo, J. Soil aggregate stability and organic matter in clay and fine silt fractions in urban refuse-amended semiarid soils. *Soil Sci. Soc. Am. J.* **2001**, *65*, 1235–1238. [CrossRef]
72. Almajmaie, A.; Hardie, M.; Doyle, R.; Birch, C.; Acuna, T. Influence of soil properties on the aggregate stability of cultivated sandy clay loams. *J. Soil Sediment* **2017**, *17*, 800–809. [CrossRef]

73. Yu, H.Y.; Ding, W.X.; Luo, J.F.; Geng, R.L.; Ghani, A.; Cai, Z.C. Effects of long-term compost and fertilizer application on stability of aggregate-associated organic carbon in an intensively cultivated sandy loam soil. *Biol. Fert. Soils* **2012**, *48*, 325–336. [CrossRef]
74. Sonnleitner, R.; Lorbeer, E.; Schinner, F. Effects of straw, vegetable oil and whey on physical and microbiological properties of a chernozem. *Appl. Soil Ecol.* **2003**, *22*, 195–204. [CrossRef]
75. Mikha, M.M.; Hergert, G.W.; Benjamin, J.G.; Jabro, J.D.; Nielsen, R.A. Long-Term Manure Impacts on Soil Aggregates and Aggregate-Associated Carbon and Nitrogen. *Soil Sci. Soc. Am. J.* **2015**, *79*, 626–636. [CrossRef]
76. Haynes, R.J.; Naidu, R.J.N.C.A. Influence of lime, fertilizer and manure applications on soil organic matter content and soil physical conditions: A review. *Nutr. Cycl. Agroecosys.* **1998**, *51*, 123–137. [CrossRef]
77. Haynes, R.J.; Beare, M.H. Aggregation and organic matter storage in meso-thermal, humid soils. In *Structure and Organic Matter Storage in Agricultural Soils*; CRC Press: Boca Raton, FL, USA, 1996; pp. 213–262.
78. Zotarelli, L.; Alves, B.J.R.; Urquiaga, S.; Boddey, R.M.; Six, J. Impact of tillage and crop rotation on light fraction and intra-aggregate soil organic matter in two Oxisols. *Soil Tillage Res.* **2007**, *95*, 196–206. [CrossRef]
79. Xie, H.T.; Li, J.W.; Zhang, B.; Wang, L.F.; Wang, J.K.; He, H.B.; Zhang, X.D. Long-term manure amendments reduced soil aggregate stability via redistribution of the glomalin-related soil protein in macroaggregates. *Sci. Rep.-UK* **2015**, *5*, 14687. [CrossRef] [PubMed]
80. Angers, D.A.; Samson, N.; Legere, A. Early Changes in Water-Stable Aggregation Induced by Rotation and Tillage in a Soil under Barley Production. *Can. J. Soil Sci.* **1993**, *73*, 51–59. [CrossRef]
81. Regelink, I.C.; Stoof, C.R.; Rousseva, S.; Weng, L.P.; Lair, G.J.; Kram, P.; Nikolaidis, N.P.; Kercheva, M.; Banwart, S.; Comans, R.N.J. Linkages between aggregate formation, porosity and soil chemical properties. *Geoderma* **2015**, *247*, 24–37. [CrossRef]
82. Wang, J.G.; Yang, W.; Yu, B.; Li, Z.X.; Cai, C.F.; Ma, R.M. Estimating the influence of related soil properties on macro- and micro-aggregate stability in Ultisols of south-central China. *Catena* **2016**, *137*, 545–553. [CrossRef]
83. Wu, X.L.; Wei, Y.J.; Wang, J.G.; Wang, D.; She, L.; Wang, J.; Cai, C.F. Effects of soil physicochemical properties on aggregate stability along a weathering gradient. *Catena* **2017**, *156*, 205–215. [CrossRef]
84. Plante, A.F.; McGill, W.B. Soil aggregate dynamics and the retention of organic matter in laboratory-incubated soil with differing simulated tillage frequencies. *Soil Tillage Res.* **2002**, *66*, 79–92. [CrossRef]
85. Demenois, J.; Rey, F.; Ibanez, T.; Stokes, A.; Carriconde, F. Linkages between root traits, soil fungi and aggregate stability in tropical plant communities along a successional vegetation gradient. *Plant Soil* **2018**, *424*, 319–334. [CrossRef]
86. Le Bissonnais, Y.; Prieto, I.; Roumet, C.; Nespoulous, J.; Metayer, J.; Huon, S.; Villatoro, M.; Stokes, A. Soil aggregate stability in Mediterranean and tropical agro-ecosystems: Effect of plant roots and soil characteristics. *Plant Soil* **2018**, *424*, 303–317. [CrossRef]