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Dynamics of water-stable aggregates associated organic carbon assessed from delta C-13 changes following temperate natural forest development in China

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ABSTRACT

In the context of global climate change, the preservation of soil productivity and the estimation of carbon budgets and cycles, the quantification of changes in carbon has important significance. In this study, we investigated the dynamics of soil aggregate associated organic carbon (OC) following temperate natural forest development in China. The objectives of this study were to examine the variation of soil aggregate associated OC decomposition rates, quantify the changes in the proportion of new and old soil aggregate OC, and explore the effects of controlling factors on SOC stocks, rate of total SOC increase and decomposition rate constants. The results showed that soil aggregate associated OC sequestration increased in 0-10 cm soil depth, while decreased in 10-30 cm soil depth. However, rate of aggregate associated OC increase, decomposition rate constants, and proportion of new OC increased at the early stage and then decreased along with the natural vegetation restoration. In addition, land use change had an important effect on soil aggregate associated OC dynamics, and soil particles, BD, MWD, C: N, plant diversity also played an important role. Moreover, SOC stocks had a negative relationship with clay and silt, while had a positive relationship with MWD and sandy soils. decomposition rate constants had a negative relationship with plant diversity, silt, and sand, while had a positive relationship with C: N and MWD. The proportions of new SOC had significant positive relationships with C: N, and it had a negative relationship with clay and silt. Therefore, it is necessary to clarify the formation mechanism of soil particles and aggregates, improve plant biodiversity, regulate the soil C: N ratio, and improve soil particle structure to increase soil carbon sequestration.

1. Introduction

In the context of global climate change and for the preservation of soil productivity the quantification of changes in carbon has important significance (Herbst et al., 2018). Conversely, land use change and climate conditions also had great effects on soil carbon pool, decomposition rate, soil aggregate stability (Deng et al., 2016; Haghighi et al., 2010), and their variation determines whether this is a carbon source or a sink. Duo to the land use change is the main driving force of global environment change, the research of soil carbon dynamics, including fixed carbon and decomposed carbon in water-stable aggregates have become an important scientific debate.

Land use strongly affects soil surface microbes, soil aggregate and soil carbon changes (van Leeuwen et al., 2017v; Zhu et al., 2018). Microbes on the outer surface of aggregates can connect neighboring aggregates through access carbon substrates (Blankinship et al., 2016). Soil aggregation affects SOC decomposition by creating complex soil structure and limiting accessibility of soil microbes to SOC (Liang et al., 2019), moreover, carbon outputs also determined by decomposition rates and turnover, influenced by climate, soil texture and plant attributes (e.g., tissue quality) (Eclesia et al., 2016). The balance of plant input and rate of SOC loss have controlled the soil carbon dynamics (Zhang et al., 2015). Generally, compared with the transformation from natural vegetation to cultivated vegetation, converting cropland into perennial vegetation had spent greater periods of time to lock up SOC due to the decrease of turnover rate related to natural vegetation (Deng et al., 2013). Thus, understanding the change in new soil aggregate associated SOC caused by new vegetation after land use change, and old

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soil aggregate associated SOC (i.e. initial soil aggregate associated SOC previous to conversation) could described more information about the dynamic responses of SOC in water-stable aggregates to land use change.

The stable carbon isotope technique can be used to quantify the SOC dynamics (Mendez-Millan et al., 2014). Conversion of vegetation regulates ¹³C in SOC because of the different isotopic signature in assimilated CO₂ as a result of photosynthetic ¹³C discrimination (Wang et al., 2015). Thus, the ¹³C value in SOC provides a means to explore the mechanisms of new and old SOC changes after land use changes (Guillaume et al., 2015; Zhang et al., 2015). Because laboratory decomposition experiments are usually combined with nutrient addition. drought, and warming, the difference in setting gradients and other experimental conditions will make the study incomparable. In-situ estimation of SOM decomposition rate makes the data obtained in various individual studies and from different regions comparable (Marin-Spiotta et al., 2009; Zhang et al., 2015). However, few researches using the stable carbon isotope technique to describe soil aggregate associated new and old SOC dynamics along a long-term vegetation restoration Chrono sequence.

Soil physic and chemical properties, land control measures, tree species planted, vegetation types and environmental factors determine the change degree of SOC stocks and the timing of the switch between increase and decrease of stocks (Arai and Tokuchi, 2010; Don et al., 2011). In addition, it is also affected by the following factors, such as interactions between geochemistry and climate (i.e., precipitation and temperature) (Doetterl et al., 2015), previous land use type (England et al., 2016; van Straaten et al., 2015v), erosion processes (Croft et al., 2012), and whether the study design is based on a chrono sequence or time series (Fujisaki et al., 2015). However, different criteria of the research blocks, such as study design and scale, stock calculation method, and land management measures, which caused the considerable uncertainties in the SOC stock dynamics (Grinand et al., 2017). For example, for forest converted into cultivated vegetation, Guo and Gifford (2002b); Don et al. (2011); Powers et al. (2011) and Fujisaki et al. (2015) given the decrease in SOC stocks ranges from 42 %, 25 %, 15.4%-8.5%, respectively. Therefore, choose to appropriately reflect the impact of overall change in soil OC stocks, and explore which factors control SOC stock dynamics is crucial to evaluate SOC change rate and its time-space distribution following land use change.

In China, the "Grain to Green" program considered as one of the most important and effective methods for the improvement of ecosystem services in the Loess Plateau (Chen et al., 2015). However, the period of these programs is too short to understand the process of ongoing vegetation restoration in Western China. Ziwuling, located in the Loess Plateau, was the only place where forest vegetation has been preserved throughout history. Thus, this area can help understand the carbon sequestration dynamics of ecosystems when converting cropland to natural restoration grassland or forest. This research used stable carbon isotope technique to investigate soil aggregate associated SOC dynamics of natural vegetation after cropland was abandoned. The vegetation had been converted from adjacent farmland for about 10, 100, 130, 150 years previously. The objective of this study were to examine the variation of soil aggregate associated SOC decomposition rates, quantify the changes in the proportion of new and old soil aggregate SOC, and explore the effects of controlling factors on SOC stocks, rate of total SOC increase and decomposition rate constant.

2. Materials and methods

2.1. Study sites

The study was conducted on the Lianjiabian Forest Farm of Ziwuling forest region of Gansu ($35^{\circ}03'$ - $36^{\circ}37'$ N, $108^{\circ}10'$ - $109^{\circ}18'$ E, 1211 - 1453 m a. s. l.), located in a total area of 23,000 km² in the hinter land of the Loess Plateau. The elevations of the hilly and gully landforms in the

Table 1			
Basic information	about	the	site.

	S1 (CK)	S2	S3	S4
Location	36°05′ N, 108°31′ E	36°02′ N, 108°31′ E	36°03′ N, 108°32′ E	36°02′ N, 108°32′ E
Slope (°) Altitude (m) Coverage (%) Shannon-Wiener index	14 1348 85 1.86	13 1450 90 2.34	10 1437 90 2.86	18 1449 95 2.59

Note: S1, Lespedeza bicolor; S2, P. davidiana; S3, P. davidiana, Q. liaotungensis; S4, Q. liaotungensis.

Table 2

Summary of features of soil and plant in different successional stages in the study areas.

	S1 (CK)	S2	S3	S4
Ρδ ¹³ C (‰)	$-24.94 \pm$	$-25.56 \pm$	$-25.53 \pm$	-25.86 ± 0.8
	0.6	0.9	1.2	
pH-a	7.57 ± 0.5	7.2 ± 0.3	7.2 ± 0.4	7.13 ± 0.6
pH-b	7.9 ± 0.4	7.57 ± 0.35	7.6 ± 0.2	7.57 ± 0.25
pH-c	7.67 ± 0.35	7.4 ± 0.2	7.03 ± 0.35	7.37 ± 0.65
Clay (%)-a	13.23 ± 0.91	9.85 ± 0.07	9.6 ± 0.28	9.21 ± 0.17
Clay (%)-b	14.32 ± 0.26	12.11 ± 0.8	12.43 ± 0.34	12.91 ± 0.51
Clay (%)-c	14.25 ± 1.16	13.31 ± 0.4	13.72 ± 0.14	13.63 ± 0.24
BD (g cm $^{-3}$)-a	1.26 ± 0.02	0.93 ± 0.02	1.03 ± 0.03	1.03 ± 0.04
BD ($g \text{ cm}^{-3}$)-b	1.24 ± 0.04	0.99 ± 0.02	1.04 ± 0.02	1.08 ± 0.06
BD ($g \text{ cm}^{-3}$)-c	1.34 ± 0.03	1.11 ± 0.06	1.25 ± 0.04	1.28 ± 0.06
Silt (%)-a	32.88 ± 2.6	27.17 ± 0.27	26.91 ± 0.14	26.86 ± 0.48
Silt (%)-b	33.69 ± 1.12	30.94 ± 0.64	32.83 ± 0.4	33.72 ± 0.73
Silt (%)-c	33.44 ± 0.08	31.38 ± 0.46	33.18 ± 0.6	35.09 ± 0.66
Sand (%)-a	53.89 ± 3.27	62.98 ± 0.33	63.49 ± 0.32	63.92 ± 0.62
Sand (%)-b	51.99 ± 1.35	56.95 ± 1.31	54.74 ± 0.12	53.38 ± 1.24
Sand (%)-c	52.31 ± 1.16	55.31 ± 0.84	53.1 ± 0.47	51.28 ± 0.86
MWD-a	3.39 ± 0.05	3.08 ± 0.09	3.25 ± 0.09	3.09 ± 0.13
MWD-b	1.52 ± 0.07	3.59 ± 0.08	3.56 ± 0.18	3.6 ± 0.08
MWD-c	0.86 ± 0.04	3.13 ± 0.15	3.34 ± 0.2	3.07 ± 0.2
TN-a	1.22 ± 0.04	2.88 ± 0.32	3.17 ± 0.22	4.29 ± 0.92
TN-b	0.62 ± 0.03	1.62 ± 0.17	0.99 ± 0.13	1.64 ± 0.11
TN-c	0.51 ± 0.04	1.19 ± 0.1	0.61 ± 0.21	0.88 ± 0.13
C: N-a	8.5 ± 0.21	12.25 ± 0.07	11.23 ± 1.21	11.06 ± 0.38
C: N-b	10.79 ± 0.31	10.24 ± 1.11	9.47 ± 1.05	9.55 ± 1
C: N-c	7.97 ± 0.36	10.35 ± 2.35	9.19 ± 1.26	8.03 ± 1.92
PN (g kg ⁻¹)-a	1.5 ± 0.48	1.44 ± 0.27	1.54 ± 0.22	1.33 ± 0.29
PN (g kg ⁻¹)-b	0.66 ± 0.00	0.91 ± 0.12	0.96 ± 0.13	0.96 ± 0.07
PN (g kg ⁻¹)-c	1.48 ± 0.00	0.99 ± 0.09	0.89 ± 0.17	0.8 ± 0.11
PC (g kg ⁻¹)-a	337.97 ±	314.34 ±	154.43 ±	379.54 ±
	34.47	1.72	28.21	43.84
PC (g kg ⁻¹)-b	325.43 ±	367.76 ±	155.54 ±	398.93 ±
	59.78	26.34	32.15	15.79
PC (g kg ⁻¹)-c	351.65 ±	359.35 ±	145.41 ±	386.71 ±
	13.09	24.73	10.18	29.52
PC: PN-a	245.15 ±	229.24 ±	$103.26 \pm$	299.58 ±
	93.75	13.72	31.21	99.85
PC: PN-b	490.11 ±	405.36 ±	162.73 ±	416.53 ±
	90.08	24.9	25.52	44.41
PC: PN-c	237.69 ±	362.43 ± 9.1	165.5 ±	487.62 ±
	8.85		23.83	41.78

Note: S1, Lespedeza bicolor; S2, P. davidiana; S3, P. davidiana, Q. liaotungensis; S4, Q. liaotungensis; $P\delta^{13}C$; plant $\delta^{13}C$; BD, soil bulk density; PC, plant carbon; PN, plant nitrogen; TN, soil total nitrogen; a, 0-10 cm; b, 10-20 cm; c, 20-30 cm soil depth.

area are 1211–1453 m.a.s.l., and their relative altitude difference is about 200 m. The area has an average annual temperature of 10 °C, an average annual rainfall of 587 mm, the accumulative temperature of 2761 °C, and the annual frost-free period is 112–140 days (Zhu et al., 2017). Most of the soil in this region is lossial, which is developed from primitive or secondary loess materials. They are evenly distributed on the red earth, with a thickness of 50-130 m, and are composed of calcareous cinnamon soil. In addition, the site is covered by a uniform



Fig. 1. Soil aggregate associated organic carbon (a, b, c and d) and MWD (e and f) for four land use types accompanying vegetation restoration. Note: The values are mean \pm SE (error bar), n = 3. MWD, mean weight diameter; S1, *Lespedeza bicolor* grassland; S2, *P. Davidiana* forest; S3, *P. davidiana* and *Q. liaotungensis* mixed forest; S4, *Q. liaotungensis* forest.

forests with abundant species, and the forest canopy density is between 80 % and 95 % (Deng et al., 2016).

them to the laboratory for air drying at room temperature.

2.2. Field investigation and sampling

Lianjiabian Forest Farm is the only remaining area of complete natural vegetation succession sequence after cultivated land (Zhang et al., 2016b). This region had been observed natural vegetation with different restoration ages, and the basic information about the site are shown in Table 1. Meanwhile, the four plots are S1 (*Lespedeza bicolor*), S2 (*P. davidiana*), S3 (*P. davidiana*, *Q. liaotungensis*), and S4 (*Q. liaotungensis*). In addition, summary of features of soil and plant in different successional stages in the study areas are shown in Table 2, and the detail forest information were described in prior studies (Deng et al., 2014b).

This study randomly set up 20 m \times 20 m plots in each forest community and 2 m \times 2 m plots in the herbaceous community to take soil samples in these five sites. The distance between two plots in a community does not exceed 50 m, and the distance between two communities in each sample area is less than 1 km. Meanwhile, this study selected consistent site conditions and land use background.

At the four corners and center of each plot in the four restoration stages, soil samples were collected at a depth of 30 cm at an interval of 10 cm with a drill bit. All soil samples of 2 mm sieved by air-dried for total OC and δ^{13} C analysis. Soil bulk density is determined by three samples with a diameter of 5 cm and a height 5 cm in each plot. The original volume of each soil core and its dry mass after oven-drying at 105 °C for 48 h were measured. Meanwhile, take 3 undisturbed soil samples in the 0 – 10, 10 – 20, 20 – 30 cm soil layers of each plot for aggregate stability analysis, seal them in a lunch boxes and transport

2.3. Sample analysis

Soil aggregates were separated by the method of first dry sieve and then wet sieve, and finally the aggregates with 6 particle sizes of > 5 mm, 2–5 mm, 1–2 mm, 0.5–1 mm, 0.25–0.5 mm and < 0.25 mm were obtained (Zhu et al., 2017). All fractions were dried at 70 °C prior to weighing. The data were analyzed to compute mean weight diameter (MWD) (Youker and McGuinness, 1957). A laser particle analyzer that operates over a range of 0.02–2000 µm (Mastersizer 2000 particle size analyzer, Malvern Instruments, Ltd., UK) and based on the laser diffraction technique was used to measure particle size. Soil pH was determined using the method of acidity agent (soil-water ration of 1:5) (PHS-3C pH acidometer, China). Soil organic carbon (SOC) content was determined by the K₂Cr₂O₇–H₂SO₄ oxidation method (Nelson and Sommers, 1996). Soil total nitrogen (TN) content was assayed using the Kjeldahl method (Bremner and Mulvaney, 1982). Fig. 1 showed the distribution of SOC in water stable aggregate.

Except δ^{13} C (in ‰ of Vienna PDB) analysis of 0.05 mm soil samples, the rest of all were at 2 mm. The natural abundance of δ^{13} C was analyzed with an Elemental Analyzer coupled to an isotope ratio mass spectrometer at the Huake Jingxin Stable Isotope Laboratory. Two acetanilide standards were measured every 12 samples. Variations in the 13 C/ 12 C ratios are reported relative to the Vienna PDB standard. The formula is expressed as:

$$\delta(\%) = (R_{sample}R_{standard-1}) \times 100 \tag{1}$$

Where R is the molar ratio of ^{13}C to ^{12}C of the sample or the international PDB reference, respectively. The $\delta^{13}C$ content data are show in



Fig. 2. Soil aggregate associated 13C for four land use types accompanying vegetation restoration. Note: The values are mean \pm SE (error bar), n = 3. S1, *Lespedeza bicolor* grassland; S2, *P. Davidiana* forest; S3, *P. davidiana* and *Q. liaotungensis* mixed forest; S4, *Q. liaotungensis* forest.

Fig. 2.

2.4. Data calculation

The proportions of new soil OC (f_{new}) and old soil OC (f_{old}) were estimated based on the mass balance formulas (Deng et al., 2016):

$$f_{new} = (\delta_{new} - \delta_{old}) \times 100\% / (\delta_{veg} - \delta_{old})$$
⁽²⁾

$$f_{old} = 100 - f_{new} \tag{3}$$

where δ_{new} is the $\delta^{13}C$ value of the soil sample from current land use, δ_{old} is the $\delta^{13}C$ values of the soil sample previous to land-use change (or soil samples from the paired 'control' sites) and d_{veg} is the $\delta^{13}C$ value of

the mixed litter of current vegetation. Decomposition rate constants (k) of soil OC were estimated using the following equations (Marin-Spiotta et al., 2009):

$$k = -\ln(C_t/C_0)/t \tag{4}$$

where C_0 is the initial soil OC stock (soil OC stock in the reference sites), C_t is initial soil OC stock remaining (old C stock) at time t (year) since land use change.

Although the increase rate of new soil C and total SOC may not be change with time since land use change, the average increase rate can be calculated using the following equation (Li et al., 2012):

Rate of increase in new soil C (or total SOC) ($g m^{-2}yr^{-1}$) = $\Delta X/\Delta t$ (5)

where Δ X is the change of new soil OC (or total SOC) stocks following land use change, and Δ t represents years since conversion (year). By multiplying the current total SOC stocks by the corresponding proportion of new soil OC, new soil OC stocks at deforestation and reforestation sites can be calculated.

Soil OC stocks were calculated using SOC concentration (g kg⁻¹), soil thickness (D, cm) and bulk density (BD, g cm⁻³) at each site (Guo and Gifford, 2002a):

$$C_s = SOC \times BD \times D/10 \tag{6}$$

The SOC sequestration was estimated using the following equation (Deng et al., 2014a):

Carbon sequestration (Mg
$$ha^{-1}$$
): $\Delta C_s = C_{LUn} - C_{LU0}$ (7)

where C_{LUn} is represent soil OC stocks at each vegetation restoration stage (g m^{-2}), and C_{LU0} is soil OC stocks at the stage of grassland.

2.5. Statistical analysis

One-way analysis of variance (ANOVA) was used to test the difference between the average values. Before performing the ANOVA procedure, all data were premised on the normality and homogeneity of variance. Correlation analysis was used to study the relationships among the different soil and plant properties. A redundancy analysis (RDA) were conducted using R program to assess the effects of soil and plant properties on soil OC stocks, the rates of total soil OC stocks increase and k increase with restoration age after farmland abandonment.

3. Results

3.1. Changes in SOC stocks and SOC sequestrations

Overall, SOC stocks and SOC sequestrations in the 0-30 cm soil depths in S2, S3, S4 were significantly higher than in S1 (Fig. 3, d and h). Except for soil aggregate (> 2 mm) associated carbon stocks and sequestrations showed significantly decreased compared to S1 in 0-10 cm soil depths (Fig. 3, c and g). SOC stocks and SOC sequestrations of S2 and S3 were the highest in the 10-30 cm soil depths, but S4 was the greatest in the 0-10 cm soil depths (Fig. 3).

In total, the rates of soil organic carbon sequestrations increased in S2, S3, and S4, however, the rates of > 2 mm soil aggregate associated carbon sequestrations decreased in 0-10 cm and 20-30 cm soil depths (Fig. 4). The rates of S3 showed the highest in 0-10 cm soil depths (), and S2 was the highest in 20-30 cm soil depths (Fig. 4).

3.2. Changes in new and old SOC

The proportion of old SOC increased, while the proportions of new SOC decreased with different restoration stage since land use change (Fig. 5, d). The gain of new SOC accounted for 30 %, 16 %, 33 % of the total SOC stocks in the S4 stage in 0-10 cm, 10-20 cm, 20-30 cm soil depths, respectively, however, the proportions of new SOC in S2 and S3 exceeded the proportions of old SOC (Fig. 5, a and b). In



Fig. 3. Soil aggregate associated organic carbon stocks (a, b, c and d) and sequestrations (e, f, g and h) for four land use types accompanying vegetation restoration. Note: The values are mean \pm SE (error bar), n = 3. S1, *Lespedeza bicolor* grassland; S2, *P. Davidiana* forest; S3, *P. davidiana* and *Q. liaotungensis* mixed forest; S4, *Q. liaotungensis* forest.

addition, the proportions of new or old SOC had different distribution trends in different soil aggregate particle size and soil depths (Fig. 5, a-c).

SOC decomposition rate decreased after S1 stages in all soil depths and soil aggregate particle size (Fig. 6). S2 had the highest SOC decomposition rate in the 10-30 cm soil depths and all soil aggregate particle size, meanwhile, S2 and S3 showed no significant difference and higher than S4 in 0-10 cm soil depths. S4 and S3 had the lowest SOC decomposition rate in 10-20 cm soil depths and 20-30 cm soil depths, respectively.



Fig. 4. Rate of total soil aggregate associated organic carbon increase for four land use types accompanying vegetation restoration. Note: The values are mean \pm SE (error bar), n = 3. S1, *Lespedeza bicolor* grassland; S2, *P. Davidiana* forest; S3, *P. davidiana* and *Q. liaotungensis* mixed forest; S4, *Q. liaotungensis* forest.

3.3. Factors effects on SOC stocks, rate of total SOC increase and decomposition rate constant

The results from RDA indicating how SOC stocks, rate of total SOC increase and decomposition rate constant relate to soil and plant properties (Fig. 7). In detail, clay, silt, TN, PN, C: N, and PC:PN played an important role in the dispersion of the sites along the first axis (Fig. 7, a). Soil aggregate associated organic carbon had a positive relationship with PN, MWD, TN, C: N, while had a negative relationship with clay, silt, BD, and PC: PN (Fig. 7, a); Plant diversity, BD, pH, PC, PC: PN, MWD, C: N and slope played an important role in the dispersion

of the sites along the first axis (Fig. 7, b). < 0.25 mm soil aggregation decomposition rate constant had a positive relationship with clay, PC: PN, PC and pH, while had a negative relationship with plant diversity, BD, slope and N. The rest of decomposition rate constant had a positive relationship with C: N, MWD, pH, PC and PC: PN, and had a negative relationship with plant diversity, BD, slope, N, and clay (Fig. 7, b); PN, clay, silt, PN, TN, C: N, PC: PN, MWD and BD played an important role in the dispersion of the sites along the first axis (Fig. 7, c). Rate of > 0.25 mm soil aggregates associated SOC increase had a positive relationship with N, C: N, plant diversity, BD and slope, while had a negative relationship with clay, silt, PC: PN, MWD and PC. The rest of rate of SOC increase had a positive relationship with N, C: N and plant diversity.

Stepwise regression analysis showed that the rate of new SOC was mainly determined by clay, decomposition rate content was mainly determined by BD, and SOC stocks were mainly determined by silt, sand, coverage and PN in the 0-10 cm soil depths (Table 3). In the 20-30 cm soil depths, the rate of new SOC was mainly determined by plant diversity and C: N, decomposition rate content and SOC stocks were mainly determined by sand (Table 3). In total, the rate of new SOC was mainly determined by Clay, plant diversity and C: N, decomposition rate content was mainly determined by SOC was mainly determined by Clay, plant diversity and C: N, decomposition rate content was mainly determined by BD, and SOC stocks were mainly determined by sand. PN and TC (Table2).

4. Discussion

4.1. Dynamic distribution of SOC stock changes

Land use change and soil depth had an effect on SOC stocks (Table 4), which was in consistent with Kirsten et al. (2018) and Qin et al. (2016). Mainly because surface soil contains most of residues and roots, and the majority of SOC change was occurred in this zone. In the process of land use change, carbon stocks of ecosystem services can be explored by plant-soil feedbacks (van der Putten et al., 2013v). Ecological succession lead to changes of in plant biomass, soil microbial community composition and other plant-soil properties (Wang et al., 2009; Zhang et al., 2016c). Therefore, the ecological successions triggered either by afforestation or by disturbances caused SOC increase (Muñoz-Rojas et al., 2015; Zhao et al., 2015), SOC decrease followed by accumulation (Menichetti et al., 2017), and limited SOC change (Bonet, 2004). In this study, soil carbon stocks and sequestrations in the 0-10cm soil depths firstly increased and then decreased along with the natural succession (Fig. 3), but in the 10-20 cm and 20-30 cm soil depths were continuously decreased (Fig. 3). These results were inconsistent with previous studies (Deng et al., 2013, 2016) in the surface soil. This is probably because: (1) higher biodiversity and functional traits increased the uptake of carbon into soil system and thus lead to higher soil carbon stocks and sequestrations (Lange et al., 2015; Steinbeiss et al., 2008); (2) stable aggregate facilitated physical protection of SOC (Six et al., 2004); (3) PC:PN had a negative relationship with soil carbon stocks (Fig. 7, a). In this study, S3 had the highest Shannon-Wiener index and the highest MWD (Tables 1 and 2), all this caused soil carbon stocks and sequestrations firstly increased and then decreased. In addition, the presence of more chemically recalcitrant compounds such as lignin and phenolics in Quercus wutaishansea than in Populus davidiana caused the rate of decomposition and carbon input were continuously decreased along with the natural succession (Wang et al., 2016; Zhong et al., 2017).

An et al. (2009); Jangid et al. (2011) and Zhang et al. (2015) found that SOC sequestration rate, soil nutrients, soil microorganisms and microbial properties all were greatest in earlier stages of restoration. These results were consistent with the rates of total SOC increase decreased in 10-20 cm and 20-30 cm soil depths in this study, but inconsistent with 0-10 cm soil depths along with the natural succession (Fig. 4). The possible mechanism was because litter biomass in S3 is the highest, so the input of new carbon increased significantly in the



Fig. 5. Proportion of new and old soil aggregate associated organic carbon for four land use types accompanying vegetation restoration (N = 3). Note: S2, P. Davidiana forest; S3, P. davidiana and Q. liaotungensis mixed forest; S4, Q. liaotungensis forest.

surface soil. Moreover, compared with S3 and S4, the low crown density and plant biodiversity of S2, was easier to cause surface carbon leaching to the deeper layer, especially in the > 2 mm soil aggregate (Fig. 4). Additionally, as vegetation succession advances, soil organic matter input and output reached a balance, so all these caused the distribution of SOC sequestration rate had a difference in the surface and deep soil.

4.2. Changes in proportions of new and old SOC following land use change

The ratio of old and new SOC can indicate the dynamics of the source of SOC in soils, and can provide important information about the dynamics of SOC after land use change (Zhang et al., 2015). In this study, the proportion of old SOC increased, while new SOC showed an opposite trend following land use change (Fig. 5). Zhang et al. (2015) and Marin-Spiotta et al. (2009) also indicated land use change was an important influencing factor to determine the proportions of new and old SOC in soils. The changes in litter quality, soil nutrient availability, and soil enzyme activity caused litter decomposition rate (Fig. 6) and new carbon input decreased following long-term vegetation recovery (Zhong et al., 2017), which produced soil organic matter with different ${}^{13}C/{}^{12}C$ ratio (Marin-Spiotta et al., 2009).

The SOC decomposition rate decreased significantly as the years of vegetation restoration increased (Fig. 6). The rate of new SOC increase was low in the early stage also because the high decomposition rate cannot contribute carbon sequestration (Marian et al., 2017). Meanwhile, the increases in old SOC in the S4 could be because the protection of soil organic matter had been restored or completely reestablished (Paul et al., 2002; Zhang et al., 2015). In addition, the rapid and bulk flow of the distribution of proportions of old SOC may bypass < 2

mm soil aggregate (Kavdır and Smucker, 2005), resulting in greater leaching to > 2 mm soil aggregate (Fig. 5).

Therefore, from the perspective of community succession, it is necessary to artificially interfere with vegetation succession to the climax community to increase carbon sequestration potential. However, from the perspective of climax community management, soil erosion should be controlled by increasing soil aggregate stability and > 2 mm soil aggregate, and thus maintaining SOC sequestration.

4.3. Factors controls over SOC stocks, rate of total SOC increase and decomposition rate constant

SOC stocks were affected by direct or indirect human induced (Smith, 2005), land use change (Don et al., 2011), soil erosion (Martinez-Mena et al., 2008), climate (Crowther et al., 2016), microbial community and activity (Huang et al., 2017), and physicochemical properties (Eze et al., 2018; Paz et al., 2016). In this study, soil particles and plant nutrients had important effects on SOC stocks, canopy also showed an influence on SOC stocks in the surface soil (Table 3 and 4). Consistent with our results (Fig. 7), Ungaro et al. (2010) and Parras-Alcántara et al. (2015) also demonstrated negative relationship between clay, silt and SOC stocks, but Kucuker et al. (2015) and Deng et al. (2016) found the opposite conclusion. The main reason is that the restoration of vegetation and proper management can increase clay, slit and SOC stocks, but the difference in sampling time, rainfall intensity or vegetation degradation lead to the reduction of clay, silt and SOC stocks. In addition, many studies have reported clay and silt content had a significant positive relationship with aggregate stability (Carter, 1992; Kemper and Koch, 1966), and < 0.25 mm aggregate associated carbon level also can influence aggregate stability (Denef et al., 2004).



Fig. 6. Soil aggregate associated organic carbon decomposition rate constants for four land use types accompanying vegetation restoration. Note: The values are mean \pm SE (error bar), n = 3. S1, *Lespedeza bicolor* grassland; S2, *P. Davidiana* forest; S3, *P. davidiana* and *Q. liaotungensis* mixed forest; S4, *Q. liaotungensis* forest.

In this study, land use change had an important effect on MWD (Table 4), and < 0.25 mm aggregate associated carbon stocks (cs1) had a positive relationship with MWD and SOC stocks (cs) (Fig. 7, a and b). Moreover, aggregate stability is one of the key parameters defining soil transportability via soil erosion (Doetterl et al., 2016), and it also had significant relationship with SOC dynamics (Bremenfeld et al., 2013). Therefore, < 0.25 mm aggregate associated carbon may cause the difference of relationships between clay content and soil carbon stocks.



Fig. 7. Redundancy analysis (RDA) of soil aggregate associated OC stocks (a), decomposition rate constants (b) and rate of new SOC increase (c), using plant and soil properties as environment variables. Note: BD, soil bulk density; Div, Shannon-Wiener index; PC, plant carbon; PN, plant nitrogen; TN, soil total nitrogen; Cs, Soil total carbon stocks; Cs1, < 0.25 mm soil aggregate associated OC stocks; Cs2, 0.25-2 mm soil aggregate associated OC stocks; Cs3, > 2 mm soil aggregate associated OC stocks; ks, Soil total carbon decomposition rate constants; ks1, < 0.25 mm soil aggregate associated OC decomposition rate constants; ks2, 0.25-2 mm soil aggregate associated OC decomposition rate constants; rc, rate of new SOC increase; rc1, rate of new < 0.25 mm soil aggregate associated OC increase; rc3, rate of new > 2 mm soil aggregate associated OC increase.

This research had a relatively low < 0.25 mm aggregate associated carbon, which caused the negative correlation between SOC stocks and clay and silt content (Fig. 7). This study also showed PN and C: N played an important role in SOC stocks (Table 3). Nitrogen availability influences the decomposition of soil organic matter, and the interaction of nitrogen and carbon offers a plethora of mechanisms to alter ecosystem carbon dynamics (Heimann and Reichstein, 2008). The C: N stoichiometry of soils also determined increases in SOC stocks (Finn et al., 2015). Therefore, the SOC stocks had positive relationship with plant and soil nitrogen and C: N ratio (Fig. 7).

Land use change, litter quality, soil properties and environmental

Table 3

Summary of stepwise regression models of measured soil variables with determining factors following vegetation restoration (N = 3).

Models	Р	R2
0–10 cm		
rc = 0.806 - 0.819 Clay	< 0.05	0.655
rc1 = 0.293 - 0.835 Clay	< 0.01	0.683
rc2 = 0.313 - 0.805 Clay	< 0.01	0.632
rc3=-0.052-0.442 Clay+0.372 C:N+0.312 Div	< 0.01	0.798
ks = 0.188 - 0.915 BD + 0.503 Clay	< 0.01	0.383
ks1=-0.01-0.731 BD + 0.461 pH	< 0.01	0.302
ks2 = 0.155-0.868 BD-0.417 Cov	< 0.05	0.403
ks3 = 0.257 - 1.05 BD - 0.648 Div	< 0.01	0.599
Cs = -5.151 + 0.663 Sand-0.385 PC:PN	< 0.01	0.799
Cs1 = 12.737 + 0.458 PN-0.614 Silt-0.327 Cov	< 0.01	0.787
Cs2 = 12.336 + 0.546 PN-0.487 Silt-0.377 Cov	< 0.01	0.757
Cs3 = 9.532 + 0.636 PN-0.545 Cov	< 0.01	0.626
10-30 cm		
rc = -0.277 + 0.936 Div + 0.173 C:N	< 0.001	0.865
rc1 = -0.162 + 0.951 Div + 0.286 C:N	< 0.001	0.834
rc2 = -0.163 + 0.987 Div + 0.259 C:N	< 0.001	0.918
rc3=-0.225 + 1.095 Div+0.217 BD	< 0.001	0.876
ks=-0.667 + 0.814 Sand	< 0.05	0.674
ks1 = 0.352 - 0.52 Silt	< 0.05	0.237
ks2 = -0.192 + 0.85 Sand $+ 0.255$ PC:PN	< 0.05	0.699
ks3 = -0.234 + 0.852 Sand	< 0.01	0.714
Cs = -6.277 + 0.552 Sand-0.448 PC + 0.376 TC	< 0.001	0.773
Cs1 = -5.672 + 0.757 Sand	< 0.01	0.553
Cs2 = -5.543 + 0.795 Sand	< 0.01	0.616
Cs3 = -6.1 + 0.432 C:N	< 0.05	0.15
0-30 cm		
rc = 0.458 - 0.692 Clay + 0.257 Div	< 0.05	0.682
rc1 = -0.09 - 0.336 Clay + 0.423 Div + 0.331 C:N + 0.230 PN	< 0.01	0.808
rc2 = -0.011 - 0.429 Clay + 0.372 Div + 0.301 C:N	< 0.01	0.734
rc3 = -0.027 - 0.397 Clay $+ 0.474$ Div $+ 0.237$ C:N	< 0.01	0.741
ks = 0.223 - 0.832 BD - 0.422 TC	< 0.01	0.415
ks1 = 0.069 - 0.729 BD + 0.533 Clay	< 0.05	0.234
ks2 = 0.044 - 0.615 BD	< 0.01	0.378
ks3 = 0.085 - 0.608 BD	< 0.01	0.37
Cs = 2.776 + 0.845 TC + 0.403 Sand + 0.132 PN	< 0.001	0.903
Cs1 = -5.074 + 0.628 Sand $+0.332$ PN	< 0.001	0.678
Cs2 = 7.007 + 0.243 PN + 0.38 Sand + 0.745 TC	< 0.001	0.708

Note: BD, soil bulk density; PC, plant carbon; PN, plant nitrogen; TN, soil total nitrogen; Cov, Covreage; Div, Shannon-Wiener index; Cs, Soil total carbon stocks; Cs1, < 0.25 mm soil aggregate associated OC stocks; Cs2, 0.25 - 2 mm soil aggregate associated OC stocks; Cs3, > 2 mm soil aggregate associated OC stocks; ks, Soil total carbon decomposition rate constants; ks1, < 0.25 mm soil aggregate associated OC decomposition rate constants; ks3, > 2 mm soil aggregate associated OC decomposition rate constants; ks3, > 2 mm soil aggregate associated OC decomposition rate constants; ks3, > 2 mm soil aggregate associated OC decomposition rate constants; ks3, > 2 mm soil aggregate associated OC decomposition rate constants; rc, rate of new SOC increase; rc1, rate of new < 0.25 mm soil aggregate associated OC increase; rc2, rate of new 0.25 - 2 mm soil aggregate associated OC increase; rc3, rate of new > 2 mm soil aggregate associated OC increase.

factors control the decomposition rate (Bronick and Lal, 2005; Knops et al., 2002). In this study, land use change had an important effect on the decomposition rate (Table 4), and it were mainly determined by soil particle size fractions and BD (Table 3). In addition, Table 4 also showed soil depth had no significant influence on soil aggregate associated OC decomposition rate, mainly because too much biomass coverage on the surface leads to consistent 0-30 cm soil temperature and humidity (Gill and Burke, 2002). Compaction can decrease porosity while increasing soil BD (Bronick and Lal, 2005), thus result in destroyed soil aggregates and then stimulate SOC decomposition rate constant as well as increasing accessibility to soil erosion (Zhou et al., 2017). Soil particles were related with diffusion of enzymes and other solutes, and diffusion is an important mechanism limiting the decomposition rate by microbial activity (Frøseth and Bleken, 2015). Based on "nitrogen mining" theory, when nitrogen is readily available in mineral form, lignin degraders have little incentive to invest resources to produce lignolytic enzymes (Zhang et al., 2016a). So even if a significant effect of tree diversity to indirectly support decomposition rate

Table 4

Two-way ANOVA results of the effects of land use type (LUC), soil depths (Dep) and their interaction (LUC \times Dep) on plant and soil properties (N = 3).

Properties	LUC (F)	LUC (P)	Dep (F)	Dep (<i>P</i>)	LUC × Dep (F)	LUC \times Dep (P)
Clay Silt Sand BD MWD pH TC TN TC TN C:N PC PN PC:PN rc rc1 rc2 rc3 ks ks1	32.282 22.732 33.1 68.193 1.003 2.006 24.318 38.11 4.243 51.464 1.192 29.478 84.229 143.973 26.507 391.097 8.239	 < 0.0001 	114.968 96.996 140.434 69.518 1.001 2.939 124.072 154.353 7.994 - - - 116.439 186.924 25.335 43.197 83.896 1.245	<pre> Comparison of the system of the sy</pre>	bec ^ Dep (F) 6.597 10.539 11.857 3.037 1 0.211 8.415 11.911 2.618 - - - 24.078 40.275 4.833 8.475 82.203 1.54	LDC × Dep (P) < 0.0001
ks2	510.57	< 0.0001	54.871	0.434	82.57	0.132
ks3	20.637	< 0.0001	2.828	0.079	2.135	0.086
Cs	20.255	< 0.0001	115.287	< 0.0001	5.332	0.001
Cs1	12.002	< 0.0001	188.758	< 0.0001	5.653	0.001
Cs2	12.561	< 0.0001	275.157	< 0.0001	9.656	< 0.0001
Cs3	1.003	0.408	0.985	0.388	1	0.448

Note: The bold data indicates significant differences in Two-way ANOVA results. BD, soil bulk density; PC, plant carbon; PN, plant nitrogen; TN, soil total nitrogen; Cs, Soil total carbon stocks; Cs1, < 0.25 mm soil aggregate associated OC stocks; Cs2, 0.25-2 mm soil aggregate associated OC stocks; Cs3, > 2 mm soil aggregate associated OC stocks; ks, Soil total carbon decomposition rate constants; ks1, < 0.25 mm soil aggregate associated OC decomposition rate constants; ks3, > 2 mm soil aggregate associated OC decomposition rate constants; ks3, > 2 mm soil aggregate associated OC decomposition rate constants; rc, rate of new SOC increase; rc1, rate of new < 0.25 mm soil aggregate associated OC increase; rc2, rate of new 0.25-2 mm soil aggregate associated OC increase.

constants by supporting the diversities of understorey herbaceous plants and soil fungi (Fujii et al., 2017), plant diversity and nitrogen also had negative relationship with decomposition rate constants (Fig. 7, b). Additionally, C: N ratio can be controlled by nitrogen addition to reduce decomposition rate constants (Zhang et al., 2016a). Meanwhile, C: N, plant diversity and clay determined the proportions of new SOC (Table 3), land use change and soil depth also influenced it (Table 4). In contrast, due to the higher rate of new SOC input into soils than new nitrogen input, the proportions of new SOC had significant positive relationships with nitrogen and C: N ratio (Fig. 7, c). The effectiveness of SOC in forming stable aggregates is also determined decomposition rate, which in turn is influenced by its physical and chemical protection from microbial action (Bronick and Lal, 2005). Thus caused C: N ratio and MWD had a positive relationship with decomposition rate constant (Fig. 7). Wang et al. (2013) showed that under similar environmental conditions, organic matter in sandy soils decomposes faster than fine-textured soils, however, the study indicated the decomposition rate constant was significantly negatively correlated with silt and sand content (Fig. 7, b). The main reason is the serious soil erosion in the study area, and SOC in the sandy soils is more likely to be lost and unstable than fine-textured soils.

In addition, consistent with results of SOC stocks (Cs), decomposition rate constants (ks) and Deng et al. (2016), land use change and soil depth influenced rate of increase of new SOC, and clay content, C: N, and plant diversity also played an important role (Tables 3 and 4). All results showed that we should improve plant biodiversity, regulate the soil C: N ratio, increase aggregate stability, and improve soil particle structure to increase soil carbon sequestration.

5. Conclusions

Land use change had a significant influence on soil aggregate associated organic carbon dynamics, and soil particles, C: N, MWD, BD and plant diversity also played an important role. Moreover, SOC stocks had a negative relationship with clay and silt, while had a positive relationship with MWD and sandy soils. decomposition rate constants had a negative relationship with plant diversity, silt, and sand, while had a positive relationship with C: N and MWD. The proportions of new SOC had significant positive relationships with C: N, and it had a negative relationship with clay and silt. Therefore, we should clarify the formation mechanism of soil particles and aggregates, improve plant biodiversity, regulate the soil C: N ratio, and improve soil particle structure to increase soil carbon sequestration. Meanwhile, based on the results of soil aggregate associated organic carbon dynamics, we also believed that it is necessary to artificially interfere with vegetation succession to the climax community to maintain high biodiversity and carbon sequestration potential.

Declaration of Competing Interest

The authors report no declarations of interest.

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