

# Decomposition characteristics of woody peat and its mixtures with labile organic materials in a newly reclaimed saline-alkali soil: A one-year incubation experiment

Yanyun ZHENG<sup>1,2</sup>, Zhenghong YU<sup>3</sup>, Jiabao ZHANG<sup>1,\*</sup>, Congzhi ZHANG<sup>1</sup>, Donghao MA<sup>1</sup>, Lin CHEN<sup>1</sup> and Guixiang ZHOU<sup>1</sup>

<sup>1</sup>State Key Laboratory of Soil and Sustainable Agriculture, Institute of Soil Science, Chinese Academy of Sciences, Nanjing 210008 (China)

<sup>2</sup>University of Chinese Academy of Sciences, Beijing 100049 (China)

<sup>3</sup>College of Agricultural Science and Engineering, Hohai University, Nanjing 211100 (China)

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## ABSTRACT

The combined application of woody peat with labile organic materials is an effective strategy for rapidly improving soil organic matter (SOM) and soil fertility in newly reclaimed lands. However, their decomposition dynamics, which can be greatly influenced by the residue-mixing effect, are not well understood. Therefore, this study investigated the decomposition characteristics and residue-mixing effect of woody peat, corn straw, organic manure, and their binary mixtures in a newly reclaimed saline-sodic soil through a 360-d incubation experiment. The results revealed that both straw and organic manure contained large amounts of O-alkyl carbon (C), leading to rapid decreases in their weight and organic C (OC) content during incubation. In contrast, the woody peat was rich in alkyl C and aromatic C, exhibiting the highest alkyl C/O-alkyl C (A/O-A) ratio and aromaticity. As a result, the woody peat only lost less than 6.4% OC and showed minimal changes in weight and total nitrogen content after one-year incubation, indicating strong degradation resistance in the saline-sodic soil. The combination of woody peat with corn straw or organic manure induced significant non-additive synergistic effects on their weight and OC losses, aromatic C, carbonyl C, A/O-A ratio, and aromaticity. This suggests that the addition of woody peat significantly slowed down the degradation of the mixtures and prolonged the retention of OC in soil. On the contrary, the mixing of corn straw and organic manure led to significant non-additive antagonistic effects on their weight and OC losses, indicating that their combination accelerated their decomposition. Consequently, this study suggests that, in contrast to the single application of various labile organic materials, the combined application of recalcitrant woody peat with labile organic materials can extend the presence of organic matter in soil, thereby promoting a sustained increase in both active and passive SOM contents and soil fertility.

**Key Words:** crop straw, newly reclaimed land, non-additive effect, organic carbon functional groups, organic manure, recalcitrant organic materials, soil fertility, soil organic matter

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## INTRODUCTION

Transforming reserved resources of cultivated lands into arable lands through soil reclamation is an important measure to alleviate concerns regarding cultivated land and food security. According to the annual Communiqué on Natural Resources of China, about  $5 \times 10^6$  ha cultivated land have been compensated from 2001 to 2017, which significantly mitigates the rapid decline of China's cultivated lands. Although the quantity of cultivated land can be expeditiously supplemented, these newly cultivated lands often suffer from limited nutrient availability, poor soil structure, low microbial activity, reduced biodiversity, and particularly low soil organic matter (SOM) content, exhibiting characteristics of inferior soil fertility and productivity (Zuo *et al.*, 2019; Xu *et al.*, 2021). Because a higher SOM content is usually associated with higher nutrient contents and microbial activities,

better soil aggregate and pore structures, more favorable soil water, heat, and air conditions, as well as stronger resistances to soil degradation (Zhang, 2023), increasing SOM content, therefore, is the key to the sustainable utilization and high productivity of agricultural soils (Yao *et al.*, 2019), especially for the newly reclaimed cultivated lands with low soil quality.

Long-term agricultural practices have shown that the application of exogenous organic materials, such as crop straw, organic manure, and green manure, is an efficient way to improve or maintain SOM content and soil productivity (Yue *et al.*, 2023). The incorporation of organic materials can stimulate soil microbial activity, thereby promoting the development of multiple organic matter pools, including particulate organic matter (POM) and mineral-associated organic matter (Angst *et al.*, 2023). Nevertheless, the buildup

\*Corresponding author. E-mail: jbzhang@issas.ac.cn.

of soil organic carbon (SOC) is a time-consuming process as most carbon (C) from these organic materials is released into the atmosphere, with only a small fraction being retained in the arable soil. For instance, Luo *et al.* (2020) reported that the SOC of a clay loam soil only increased slightly by 7.1%–15.4% (from 12.3–13.2 to 13.6–14.9 g kg<sup>-1</sup>) after continuous application of crop straw at a rate of 5 Mg ha<sup>-1</sup> year<sup>-1</sup> over 30 years. Obviously, such a high time cost is not conducive to the desire for the rapid and efficient utilization of newly reclaimed cultivated lands, if only labile exogenous organic materials are applied.

Generally, according to the stability or turnover time of organic C in soil, the SOC pool can be divided into the active SOC pool (labile pool) with a short turnover time of days to months and the passive SOC pool (non-labile pool) with a much longer turnover time of years to centuries (Parton *et al.*, 1987; Chen Z D *et al.*, 2019). The active SOC, which is usually present in the soil as free POM and dissolved organic matter, is the key to soil productivity and the most important and direct nutrient and energy source for soil microbial activities (Angst *et al.*, 2023). In contrast, the passive SOC, which is usually protected against microbial process through molecular recalcitrance, physical occlusion, or chemical interaction with mineral surface, is a vital assurance for the sustained supply of nutrients for crop growth and microbial activity, as well as for maintaining an optimal soil structure and a high soil fertility level (Lützow *et al.*, 2006; Lal, 2023). In fertile arable soils, the active SOC pool ordinarily accounts for 10%–50% of the total SOC pool, while the passive SOC pool can approach about 50%–90% (Just *et al.*, 2023). Clearly, to quickly increase the SOM content and soil fertility of the newly reclaimed cultivated lands, it is imperative to not only replenish the active SOC by applying labile organic materials, but also augment the passive SOC pool through the incorporation of recalcitrant organic materials.

Woody peat, a transitional substance resulting from the natural transformation of woody biomass into lignite over the course of tens of thousands of years, is characterized by abundant humus and strong water retention and nutrient preservation capacities (Limpens *et al.*, 2008). Humus, which consists of decomposed organic matter or its re-synthesized by-products, is usually used as a synonym for SOM in a narrow sense (Dou *et al.*, 2020). Our previous study suggested that the humus content of woody peat can be as high as 860 g kg<sup>-1</sup>, which is 2–3 times that of organic fertilizer and 15–40 times that of soil (Zheng *et al.*, 2018). In addition, the C/nitrogen (N) ratio of woody peat can approach about 70, and its molecular structure contains more aromatic functional groups than organic fertilizer (Zheng *et al.*, 2018). These indicate that woody peat has a more complex and recalcitrant molecular structure and therefore can be used to swiftly

increase SOM content and soil fertility. Afterwards, a lot of studies have been conducted to investigate the effects of woody peat on SOM, soil structure, microbial and enzyme activities, and soil fertility and productivity. Chang *et al.* (2019) and Yu *et al.* (2023) reported that the addition of woody peat can help to reduce the losses of SOC and N, improve root activity, and promote nutrient uptake by roots. Fu *et al.* (2021) and Zhao *et al.* (2023) showed that the application of woody peat can significantly increase the SOM contents and crop yields of different degraded or newly reclaimed lands. Xuan *et al.* (2022) showed that woody peat had a better improvement effect on soil macro porosity than maize straw. Particularly, Li *et al.* (2021), Zhang *et al.* (2021), and Duan *et al.* (2023) suggested that the combined application of woody peat and labile organic materials, such as straw and organic fertilizer, can lead to higher SOM contents and crop yields compared to woody peat alone. This result could be partially explained by the average mixing effect or the additive effect of organic materials with different decomposition rates. Yet, the combination of organic materials of distinct chemical compositions and decomposition capacities often gives rise to non-additive effects, which may enhance or inhibit the decomposition of the mixtures (Zhou *et al.*, 2019). Despite this, our knowledge of the decomposition characteristics of woody peat and its mixtures with labile organic materials in newly reclaimed lands remains limited, especially in terms of the non-additive effects resulting from the mixing of woody peat with labile organic materials.

Therefore, the objective of this study was to investigate the decomposition characteristics of woody peat and its mixtures with crop straw or organic manure in a typical newly cultivated saline-alkali soil through a one-year litterbag incubation experiment. Because of the extreme differences in chemical composition and molecular recalcitrance among woody peat, crop straw, and organic manure, we hypothesized that the combination of woody peat with labile organic materials may not only lead to an average mixing effect but also result in significant non-additive effects, potentially leading to a prolonged retention of the woody peat mixtures in a saline-alkali soil. The reason for selecting this soil is that there are around  $3.4 \times 10^7$  ha of saline-alkali lands in China, among which more than 36% can be potentially utilized for agricultural production after reclamation (Wang *et al.*, 2017).

## MATERIALS AND METHODS

### *Soil and organic materials*

The saline-sodic soil used in this study is classified as Haplic Solonetz according to the World Reference Base for Soil Resources Bank (FAO/ISRIC/ISSS, 1998). It was

collected from a newly reclaimed cultivated land (35°23'31" N, 118°42'51" E) located in Xinxing Town, Tongyu County, Baicheng City, Jilin Province, China. The area has a northern temperate continental monsoon climate with a mean annual temperature of 5.2 °C and a mean annual precipitation of 408 mm. The land was reclaimed in April 2016. Soil sample was collected from the plough horizon (0–20 cm) in June 2016, air-dried, and sieved through a 2-mm sieve. The basic physicochemical characteristics of the soil, determined according to the methods listed in Table SI (see Supplementary Material for Table SI), are shown in Table SII (see Supplementary Material for Table SII). The woody peat was provided by the Zhongxiang Lifeng Technology Co., Ltd., Beijing, China. The corn straws had been oven-dried at 60 °C and cut to a length of less than 0.5 cm. The organic manure, purchased from the Lianye Biological Technology Co., Ltd., Jiangsu, China, was produced by fermenting a mixture of livestock and poultry manure, corn straw, and soybean meal. The chemical properties of these three organic materials, determined using the methods listed in Table SI, are summed in Table SIII (see Supplementary Material for Table SIII).

#### *Litterbag incubation experiment*

The litterbag incubation method was used to investigate the decomposition characteristics of woody peat and its mixtures with labile organic materials in the saline-sodic soil. Six treatments were designed as follows: i) woody peat (wP), ii) corn straw (cS), iii) organic manure (oM), iv) woody peat + corn straw (PS), v) woody peat + organic manure (PM), and vi) organic manure + corn straw (MS). Due to the large volume of corn straw, only 4 g of corn straw was used in each relevant treatment, while 8 g of wet woody peat and 8 g of wet organic manure were used. The reason for using wet woody peat and organic manure was to avoid altering their microbial communities and physicochemical activities. The dry weights of the woody peat and organic manure were measured after oven-drying at 100 °C and used to calculate the weight losses in relevant treatments.

For each treatment, the above organic materials were weighed, mixed thoroughly, and put into a nylon litterbag (8 cm × 10 cm, 0.013-mm mesh). Twelve bags were prepared for each treatment. Then, a sealed litterbag was horizontally buried in the center of a rectangular plastic box containing 750 g of the air-dried saline-sodic soil. The thickness of the soil below and above the litterbag was 2 and 4 cm, respectively. Afterwards, all boxes were placed in the dark for 360 d at a constant temperature of 25 °C. During the entire incubation period, soil moisture was maintained at 70% water-holding capacity by the weighing method. The water-holding capacity of the saline-sodic soil was determined using the Wilcox method (Gao *et al.*, 2016) to be 31.0%. After 30, 90, 180, and 360 d of incubation, three litterbags

were retrieved from each treatment, and the adhered soils were gently wiped off. The residual organic material within each litterbag was sampled for the determination of microbial community structure. The remaining was oven-dried at 60 °C, weighed, and ground to < 0.25 mm for further analysis. According to the weight differences before incubation (day 0) and after 30, 90, 180, and 360 d of incubation, the weight loss percentages ( $W_t$ , %) of the organic materials were calculated.

#### *Chemical analysis*

Organic C (OC) contents of the organic materials at different decomposition stages were measured by oxidation with potassium dichromate (Lu, 2000). Total N (TN) content was determined with an elemental analyzer (Elementar, Germany). The ratio of C/N was calculated based on the OC and TN contents. For the determination of total phosphorus (TP) and potassium (TK) contents, organic materials were digested using H<sub>2</sub>SO<sub>4</sub>-H<sub>2</sub>O<sub>2</sub>. Then, TP was analyzed using the molybdenum-antimony-ascorbic acid spectrophotometric method, and TK was determined using a flame photometer (Shanghai Jingke, China) (Lu, 2000).

#### *Chemical structural composition determination*

Chemical structural compositions of the organic materials in different treatments before incubation (day 0) and after 90, 180, and 360 d of incubation were determined by solid-state <sup>13</sup>C cross-polarization (CP)/total sideband suppression (TOSS) nuclear magnetic resonance (NMR) spectroscopy. Although the CP technique often underestimates the aromatic C, it can still give useful semi-quantitative information about the organic functional groups (Quideau *et al.*, 2000; Mao and Schmidt-Rohr, 2004; Gerke, 2018). The 30-d samples were not determined due to the too short time interval. The <sup>13</sup>C NMR analysis was carried out on a Bruker Advance III 600 spectrometer (Bruker, Germany) operating at a <sup>13</sup>C resonance frequency of 100.6 MHz with a 4-mm zirconia rotor. The spinning speed was 5 kHz, and a contact time of 10 ms, a spectral width of 100 kHz, a 90° pulse width of 4 μs, a cycle delay of 1 s, and a scan rate of 4 096 h<sup>-1</sup> were used. The chemical shift was calibrated with the standard substance glycine. The obtained <sup>13</sup>C CP/TOSS NMR spectra were subdivided into four chemical shift regions and assigned to the following organic C functional groups (Wang *et al.*, 2015): alkyl C (chemical shift (δ) = 0–50), O-alkyl C (δ = 50–110), aromatic C (δ = 110–160), and carbonyl C (δ = 160–200). The relative abundance of each functional group was obtained by integrating and normalizing the spectral area to the total signal intensity (δ = 0–200) for each spectrum. The alkyl C/O-alkyl C (A/O-A) ratio was calculated as the ratio of the relative abundance of alkyl C to that of O-alkyl C. Aromaticity was calculated as the relative abundance of

aromatic C divided by the sum of the relative abundances of alkyl C, O-alkyl C, and aromatic C. The A/O-A ratio is a sensitive index of the degree of decomposition, and aromaticity is an important index of humification (Xu *et al.*, 2019).

### Residue-mixing effect analysis

Due to the varying quality of different materials, the decomposition rate of an organic mixture can deviate greatly from the expected rate based on the additive decomposition of the mixture (Hoorens *et al.*, 2003). The decomposition may be either faster or slower than expected because of the interaction between different organic materials or the residue-mixing effect. Hence, to evaluate the residue-mixing effects on organic material weight loss, OC, TN, TK, TP, organic functional groups (alkyl C, O-alkyl C, aromatic C, and carbonyl C), A/O-A ratio, and aromaticity, the strength of possible interactions (SPI) was calculated as follows (Zhou *et al.*, 2019):

$$SPI = \frac{OBS \text{ value}}{EXP \text{ value}} - 1 \quad (1)$$

where OBS value is the determined value of  $W_t$ , OC, TN, TK, TP, relative abundances of organic functional groups, A/O-A ratio, or aromaticity of the residual organic material in a mixture treatment (PS, PM, or MS), and EXP value is the average  $W_t$  of the respective single organic material treatments (wP and cS, wP and oM, or oM and cS, respectively), which was calculated as follows (Hoorens *et al.*, 2003):

$$EXP \text{ value} = n_1M_1 + n_2M_2 \quad (2)$$

where  $M_1$  and  $M_2$  are the determined values of  $W_t$  of the 1st and 2nd organic materials, respectively, and  $n_1$  and  $n_2$  are their weight percentages in the mixture, respectively. One-sample *t*-test was used to analyze whether SPI was significantly different from zero. If SPI is significantly larger than 0, it indicates positive (synergistic) residue-mixing effects between the two organic materials on the weight loss and properties (OC, TN, TK, TP, relative abundances of organic functional groups, A/O-A ratio, and aromaticity) of the mixture, otherwise, it indicates negative (antagonistic) residue-mixing effects (Zhou *et al.*, 2019). The greater the deviation from zero, the stronger the synergistic or antagonistic effects.

### Statistical analysis

One-way analysis of variance was performed to determine the difference in chemical composition among treatments, and the significance of the difference between means was tested by the least significant difference method at  $P < 0.05$ . The strength of the residue-mixing effect was evaluated using one-sample *t*-test. All statistical analyses were

performed using IBM SPSS Statistics 20. The heatmaps of correlation coefficients were generated using Pearson correlation in Origin 2021. Principal component analysis (PCA) was carried out on the variance-covariance matrix of soil properties and functional group data of days 0, 90, 180, and 360 of different treatments using IBM SPSS Statistics 20.

## RESULTS

### Organic material weight loss during incubation

The organic material  $W_t$  in different treatments during the 360-d incubation are shown in Fig. 1. The results suggest that the organic material weight in wP barely changed during the whole incubation period in the saline-sodic soil. In contrast, the rapid organic material weight loss in cS indicated the fastest decomposition rates (31.3%, 47.0%, 59.0%, and 66.9% at days 30, 90, 180, and 360, respectively) throughout the incubation period, followed by those in oM (0.6%, 7.2%, 13.6%, and 14.5%, respectively). Compared to cS, adding woody peat in PS resulted in much lower  $W_t$  values of 15.0%, 15.8%, 23.5%, and 30.4% at days 30, 90, 180, and 360, respectively. Compared to oM, adding woody peat in PM resulted in much lower  $W_t$  values of 2.6%, 5.8%, and 9.5% at days 90, 180, and 360, respectively. However, the mixture of organic manure and corn straw (MS) showed high  $W_t$  values (only second to those of cS) during the whole incubation period. The  $W_t$  values of different treatments were in the descending order of cS > MS > PS > oM > PM > wP.

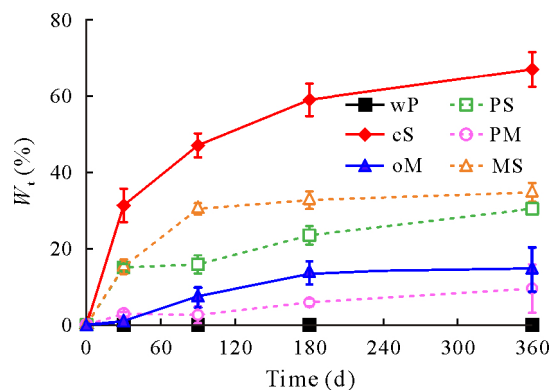


Fig. 1 Organic material (OM) weight loss percentages ( $W_t$ ) in the different decomposition treatments with OM-containing litterbags buried in a newly reclaimed saline-sodic soil and incubated at 25 °C for 360 d. Error bars are standard deviations of the means ( $n = 3$ ). wP = woody peat; cS = corn straw; oM = organic manure; PS = woody peat + corn straw; PM = woody peat + organic manure; MS = organic manure + corn straw.

### Organic material nutrient content changes during incubation

After 360 d of incubation, the organic material OC contents in all treatments declined as compared to day 0 (Fig. 2). However, there were significant differences in decrease magnitude among the treatments. The woody peat-containing

treatments (wP, PS, and PM) showed much smaller OC content decreases (only 5.7%, 2.5%, and 5.5%, respectively) than oM, cS, and MS where decreases of 36.1%, 15.5%, and 32.6%, respectively, were observed. According to Figs. 1 and 2, the woody peat only lost less than 6.4% OC after 360 d of incubation. The changes in TN content for different treatments were greatly different from those in OC content. Before the incubation (day 0), oM had the highest TN content while cS had the lowest. However, after only 30 d, the TN content in oM showed the most significant decrease (27.9%) among the different treatments. On the contrary, the TN content in cS increased more than 4-fold after 360-d of incubation. Due to the residue-mixing effect, the mixture of organic manure and corn straw (MS) exhibited an increasing trend in TN content. In contrast, the TN content in wP

showed the smallest change after 360 d of incubation. When woody peat was added, the drastic changes in TN content in cS and oM were greatly attenuated. The C/N ratio decreased significantly during the 360-d incubation in all treatments except PM (Table I). From day 30 to 360, the C/N ratios in wP, PS, and PM showed small decreases (10.6%–26.1%) to still higher than 25, while the C/N ratios in cS, oM, and MS were greatly decreased by 72.2%, 38.5%, and 34.6%, respectively.

The TP and TK contents in all treatments showed rapid decrease trends (Fig. 3). Due to the much higher contents of TP and TK in the organic manure compared to the woody peat and corn straw, oM, MS, and PM showed higher TP and TK contents than wP, cS, and PS during the whole incubation period, with wP exhibiting the lowest TP and

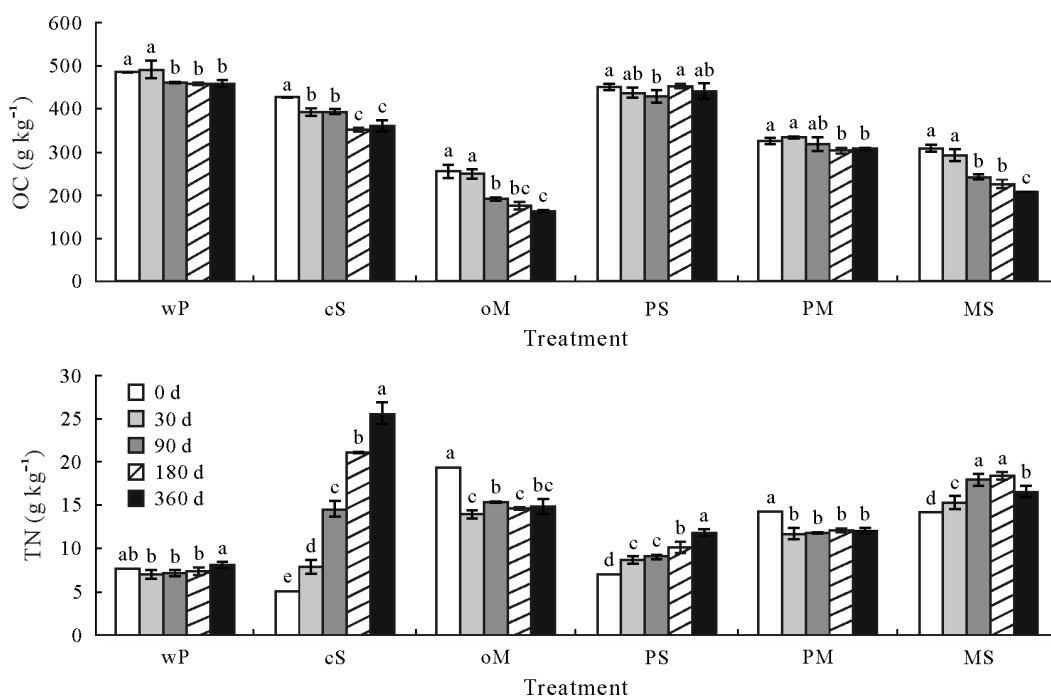


Fig. 2 Organic C (OC) and total N (TN) contents in organic materials (OM) in the different decomposition treatments with OM-containing litterbags buried in a newly reclaimed saline-sodic soil and incubated at 25 °C for 360 d. Error bars are standard deviations of the means ( $n = 3$ ). Different letters indicate significant differences between times for the same treatment at  $P < 0.05$ . wP = woody peat; cS = corn straw; oM = organic manure; PS = woody peat + corn straw; PM = woody peat + organic manure; MS = organic manure + corn straw.

TABLE I

C/N ratio changes in organic materials (OM) in the different decomposition treatments with OM-containing litterbags buried in a newly reclaimed saline-sodic soil and incubated at 25 °C for 360 d

Treatment <sup>a)</sup>	0 d	30 d	90 d	180 d	360 d
wP	62.93 ± 0.13 <sup>b)c)</sup>	69.45 ± 3.53a	64.86 ± 3.21ab	62.67 ± 2.83b	56.52 ± 2.44c
cS	84.89 ± 0.40a	50.67 ± 7.26b	26.99 ± 1.66c	16.65 ± 0.17d	14.09 ± 0.20d
oM	13.27 ± 0.76b	17.98 ± 1.31a	12.46 ± 0.35bc	12.10 ± 0.62bc	11.05 ± 0.56c
PS	64.39 ± 0.80a	50.72 ± 1.25b	47.09 ± 0.77c	45.11 ± 2.62c	37.46 ± 2.33d
PM	22.84 ± 0.42c	28.61 ± 1.81a	27.05 ± 1.56ab	25.18 ± 0.47b	25.57 ± 0.69b
MS	21.83 ± 0.53a	19.24 ± 1.27b	13.34 ± 0.28c	12.31 ± 0.38c	12.58 ± 0.55c

<sup>a)</sup>wP = woody peat; cS = corn straw; oM = organic manure; PS = woody peat + corn straw; PM = woody peat + organic manure; MS = organic manure + corn straw.

<sup>b)</sup>Mean ± standard deviation ( $n = 3$ ).

<sup>c)</sup>Different letters in the same row indicate significant differences between times at  $P < 0.05$ .

TK contents. Due to the residue-mixing effect, PS and PM showed intermediate TK and TP contents between wP and cS or oM.

*C chemistry of organic materials during incubation*

Several organic C functional group signals with different

intensities at  $\delta = 29, 55, 72, 103, 128, 145,$  and  $172$  were recognized in the  $^{13}\text{C}$  CP/TOSS NMR spectra of the organic materials before incubation (day 0) and after 90, 180, and 360 d of incubation (Fig. 4). The signal at  $\delta = 29$  belonged to alkyl C ( $\delta = 0\text{--}50$ ), which is usually attributed to polymethylene, a constituent of lipid substances such as

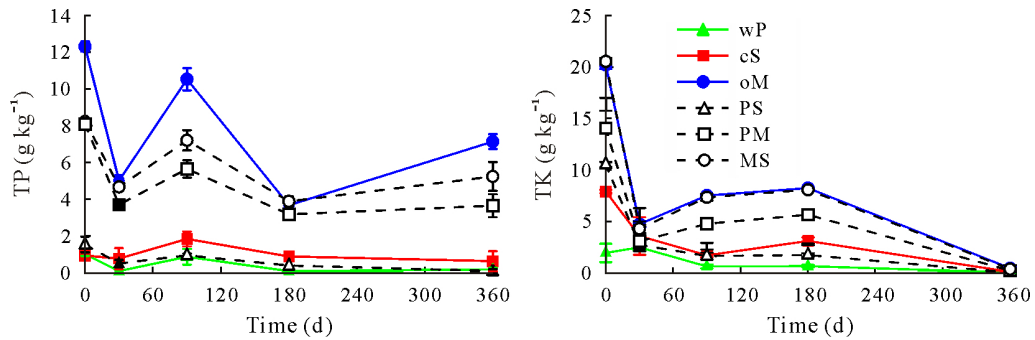


Fig. 3 Total P (TP) and K (TK) contents in organic materials (OM) in the different decomposition treatments with OM-containing litterbags buried in a newly reclaimed saline-sodic soil and incubated at 25 °C for 360 d. Error bars are standard deviations of the means ( $n = 3$ ). wP = woody peat; cS = corn straw; oM = organic manure; PS = woody peat + corn straw; PM = woody peat + organic manure; MS = organic manure + corn straw.

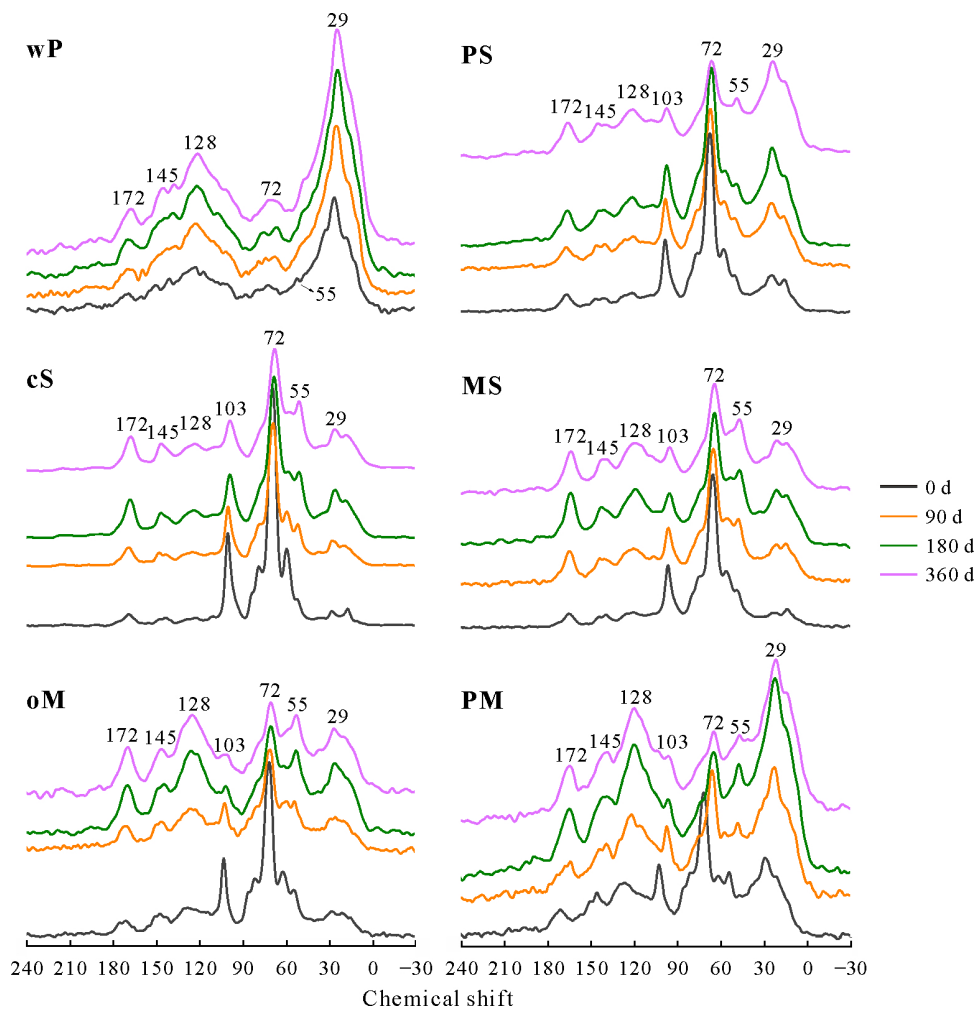


Fig. 4 Solid-state  $^{13}\text{C}$  cross-polarization/total sideband suppression nuclear magnetic resonance spectra of organic materials (OM) in the different decomposition treatments with OM-containing litterbags buried in a newly reclaimed saline-sodic soil and incubated at 25 °C for 360 d. wP = woody peat; cS = corn straw; oM = organic manure; PS = woody peat + corn straw; MS = organic manure + corn straw; PM = woody peat + organic manure.

cutin, suberin, and waxes (Alexis *et al.*, 2012; Wang *et al.*, 2015). The signals at  $\delta = 55$ , 72, and 103 were assigned to O-alkyl C ( $\delta = 50$ –110), which typically derives from methoxy C in lignin (Alexis *et al.*, 2012; Panettieri *et al.*, 2014), saccharide compounds such as cellulose, hemicellulose, and carbohydrates (Jindo *et al.*, 2011; Mao *et al.*, 2011), and sugar rings of polysaccharides (Zhou *et al.*, 2014), respectively. The signals at  $\delta = 128$  and 145 can be assigned to aromatic C ( $\delta = 110$ –160) (Alexis *et al.*, 2012). Finally, the signal at  $\delta = 172$  belonged to carbonyl C ( $\delta = 160$ –200), which generally indicates carboxylate, peptides, and amides (Alexis *et al.*, 2012; Zhou *et al.*, 2014). Based on the NMR spectra of different treatments, the relative abundances of alkyl C ( $\delta = 0$ –50), O-alkyl C ( $\delta = 50$ –110), aromatic C ( $\delta = 110$ –160), and carbonyl C ( $\delta = 160$ –200) were calculated (Fig. 5).

Before the incubation (day 0), the NMR spectrum of the woody peat (wP) exhibited three intense signals at  $\delta = 29$ , 128, and 145 and two relatively weak signals at  $\delta = 72$  and 172 (Fig. 4), which indicated that the woody peat was mainly composed of alkyl C (45.6%) and aromatic C (26.3%) (Fig. 5). Besides, during the whole incubation, there was no significant change in the signal intensities and relative abundances of the four functional groups in wP. In contrast, the NMR spectra of cS and oM showed intensive peaks at  $\delta = 72$  and 103 on day 0, suggesting high relative abundances of O-alkyl C in both corn straw and organic manure, which were 82.6% and 62.0%, 3.8 and 2.8 times that of woody peat, respectively. On the contrary, before the incubation (day 0), the total abundances of alkyl C and aromatic C in cS and oM were only 14.6% and 33.3%, being merely 20.3% and 46.3%, respectively, of those in wP. During the incubation period, the signal intensity and relative abundance of O-alkyl C showed rapid and drastic decreases in both cS and oM, which led to relative increases in the relative abundances of alkyl C and aromatic C. Furthermore, compared to corn straw (cS), organic manure (oM) showed stronger signals of alkyl C ( $\delta = 29$ ) and aromatic C ( $\delta = 145$  and 128) before

incubation (day 0) and a weaker signal of O-alkyl C ( $\delta = 72$ ) after incubation (day 360), which indicated that organic manure contained relatively more recalcitrant compounds. When organic manure and corn straw were mixed (MS), the relative abundance of O-alkyl C remained above 70%, and even after 360 d of incubation, it was still more than 40%. On the contrary, mixing woody peat with corn straw or organic manure (PS and PM) could immediately increase the total relative abundances of alkyl C and aromatic C to 31.1% and 49.8%, respectively, which were 1.5–2.3 times those in oM and cS. After 360 d of incubation, the relative abundances of alkyl C in PS and PM increased to 35.2% and 37.1%, respectively.

According to the relative abundances of alkyl C ( $\delta = 0$ –50), O-alkyl C ( $\delta = 50$ –110), aromatic C ( $\delta = 110$ –160), and carbonyl C ( $\delta = 160$ –200) shown in Fig. 5, the A/O-A ratio and aromaticity were calculated. Before the incubation, both the A/O-A ratio and aromaticity of woody peat (wP) were the highest among the different treatments (Fig. 6). In contrast, at the beginning of incubation (day 0), the A/O-A ratio and aromaticity of corn straw (cS) were only 4.9% and 23.0% of those in wP, respectively, and the A/O-A ratio of organic manure (oM) was merely 10.9% of that in wP. Meanwhile, there was no significant change in these two parameters in wP during the incubation period. Different from wP, the A/O-A ratio and aromaticity in cS and oM showed marked increases during the incubation. Adding woody peat to either corn straw or organic manure resulted in a much higher A/O-A ratio and aromaticity than in the corresponding organic material without addition. Besides, greater increases in the A/O-A ratios in PS and PM during the incubation were observed compared to cS, oM, and MS.

#### *Relationships between decomposition rate and chemical properties of organic materials*

The correlation coefficients between  $W_t$  and the contents of OC, TN, TK, and TP, and C/N ratio showed that  $W_t$

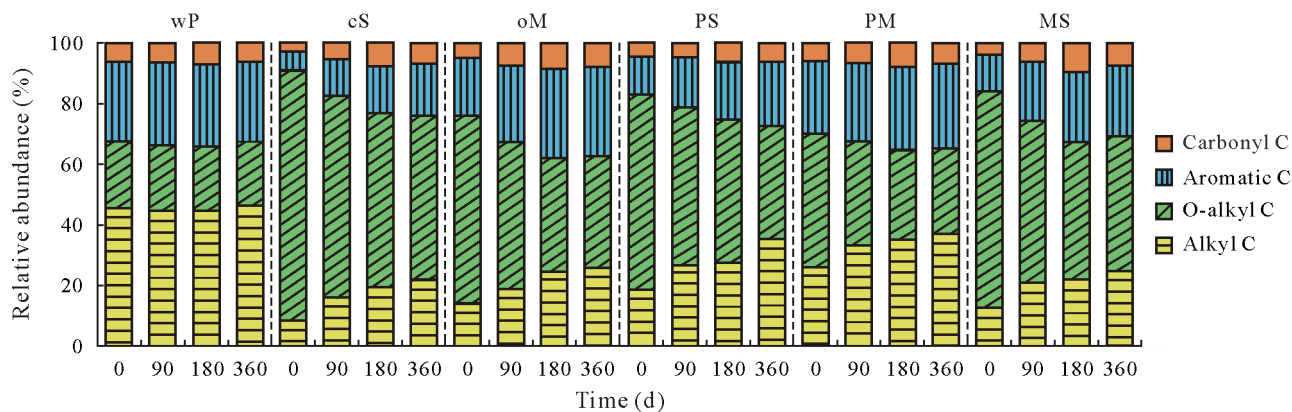


Fig. 5 Relative abundances of organic C functional groups in organic materials (OM) in the different decomposition treatments with OM-containing litterbags buried in a newly reclaimed saline-sodic soil and incubated at 25 °C for 360 d. wP = woody peat; cS = corn straw; oM = organic manure; PS = woody peat + corn straw; PM = woody peat + organic manure; MS = organic manure + corn straw.

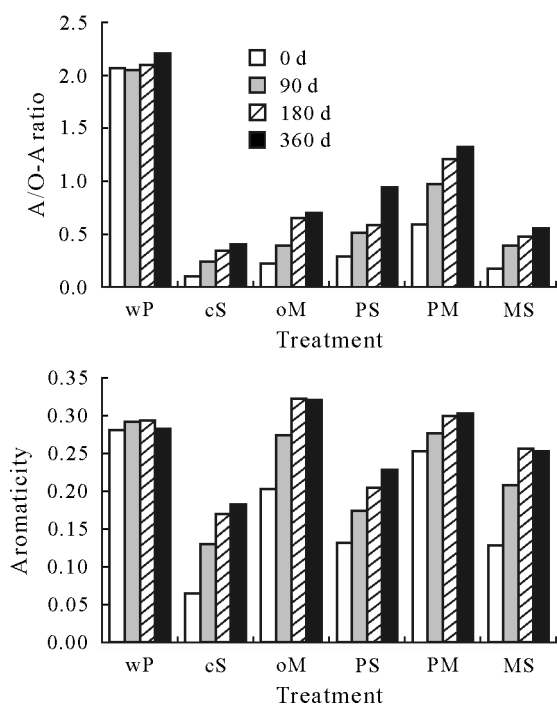


Fig. 6 Alkyl C/O-alkyl C (A/O-A) ratios and aromaticity of organic materials (OM) in the different decomposition treatments with OM-containing litterbags buried in a newly reclaimed saline-sodic soil and incubated at 25 °C for 360 d. wP = woody peat; cS = corn straw; oM = organic manure; PS = woody peat + corn straw; PM = woody peat + organic manure; MS = organic manure + corn straw.

had a significant positive correlation with TN content in all treatments except oM, and it had a significant negative correlation with C/N ratio in all treatments except PM (Fig. 7). This was probably due to the high TN contents of the organic materials (Table SIII). In wP and PS,  $W_t$  was significantly negatively correlated with TK content, which indicated that potassium may also be an important factor in these two treatments. The correlation coefficients between  $W_t$ , OC,

TN, C/N ratio, TK, TP, and the relative abundances of alkyl C, O-alkyl C, aromatic C, and carbonyl C after 90, 180, and 360 d of incubation showed that during the whole incubation,  $W_t$  was significantly positively correlated with O-alkyl C but negatively correlated with alkyl C and aromatic C (Fig. 8). The OC content showed significant negative correlations with TK and TP contents and a negative correlation with carbonyl C during the entire incubation. Besides, TN content showed significant negative correlations with alkyl C and aromatic C but a positive correlation with O-alkyl C.

*PCA of chemical composition and organic C functional groups of organic materials*

The PCA analysis was applied to assess the factors that influenced the decomposition of different organic materials. The eigenvalues are presented in Table SIV (see Supplementary Material for Table SIV). The 1st (PC1) and 2nd (PC2) principal components explained 47.3% and 34.8% of the total variance, respectively (Fig. 9). The plot of the PC scores showed clear differences among different treatments, suggesting distinct decomposition capacities across treatments (Fig. 9a). Almost all treatments exhibited increasing trends along both PC1 and PC2 with incubation time, indicating that higher PC1 and PC2 scores corresponded to greater degrees of decomposition. Throughout the incubation period, the PC1 scores of woody peat (wP) were consistently larger than those of corn straw (cS) and organic manure (oM), and the differences between sampling times were also much smaller in wP, suggesting a much slower decomposition rate for woody peat. The addition of woody peat obviously increased the scores of corn straw and organic manure along PC1 and decreased the difference between sampling times along PC2. The factor loadings of the parameters suggested

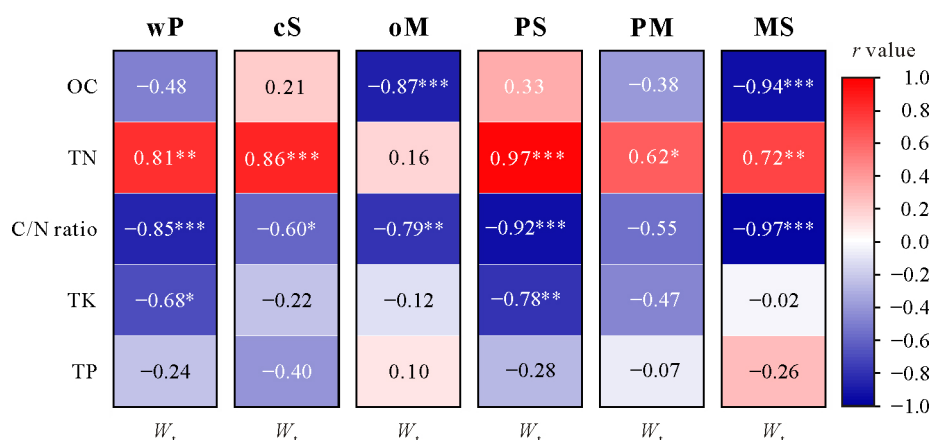


Fig. 7 Heatmaps of the correlation coefficients ( $r$ ) between weight loss percentage ( $W_t$ ) and organic C (OC), total N (TN), total K (TK), and total P (TP) contents and C/N ratio of organic materials (OM) in the different decomposition treatments with OM-containing litterbags buried in a newly reclaimed saline-sodic soil and incubated at 25 °C for 360 d. Asterisks \*, \*\*, and \*\*\* indicate significances at  $P < 0.05$ ,  $P < 0.01$ , and  $P < 0.001$ , respectively. wP = woody peat; cS = corn straw; oM = organic manure; PS = woody peat + corn straw; PM = woody peat + organic manure; MS = organic manure + corn straw.

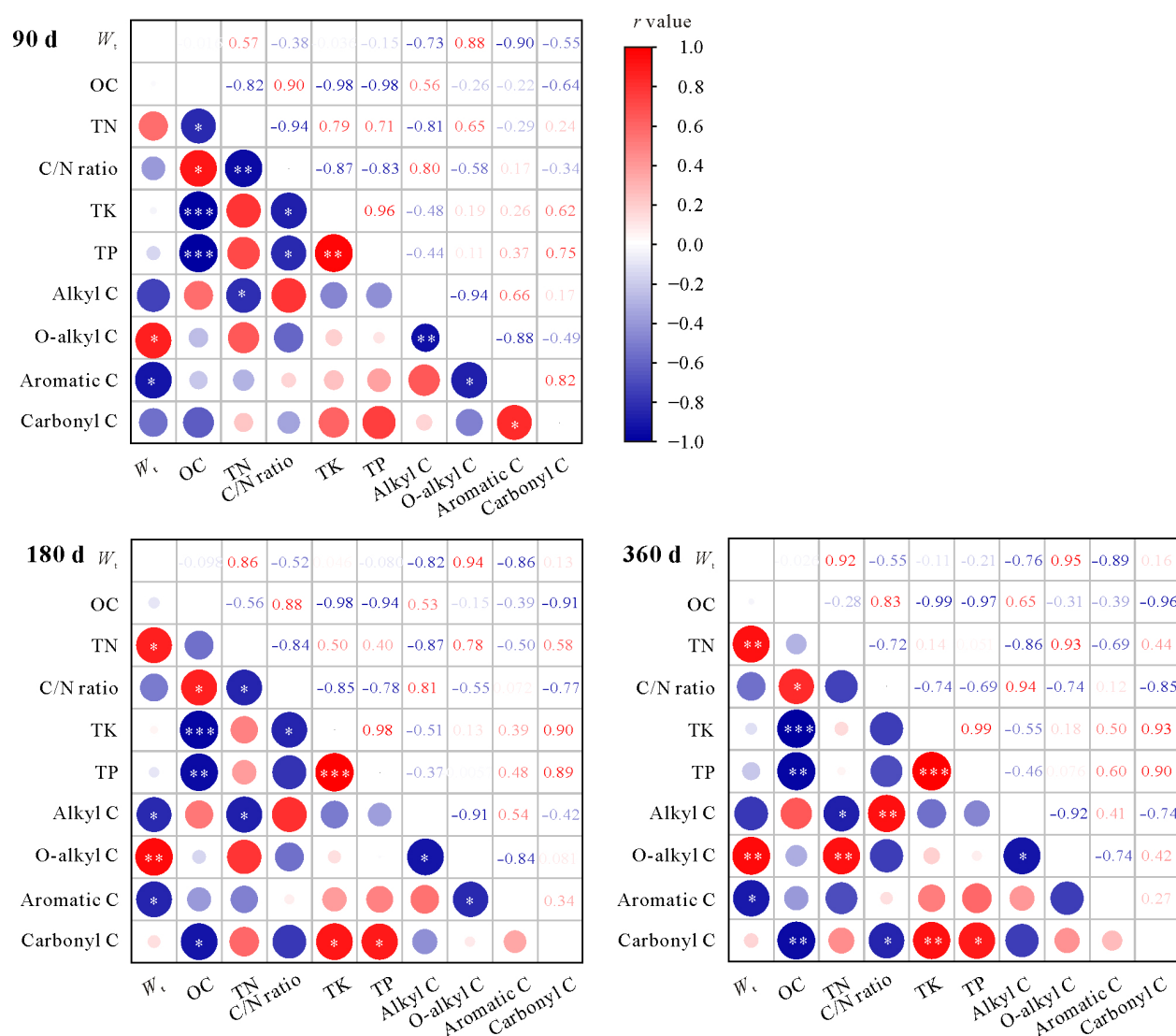


Fig. 8 Heatmaps of the correlation coefficients ( $r$ ) between weight loss percentage ( $W_t$ ), organic C (OC), total N (TN), total K (TK), and total P (TP) contents, C/N ratio, and the relative abundances of alkyl C, O-alkyl C, aromatic C, and carbonyl C of organic materials (OM) in a decomposition experiment with OM-containing litterbags buried in a newly reclaimed saline-sodic soil and incubated at 25 °C for 360 d. Asterisks \*, \*\*, and \*\*\* indicate significances at  $P < 0.05$ ,  $P < 0.01$ , and  $P < 0.001$ , respectively.

that aromatic C, aromaticity, alkyl C, A/O-A ratio, and carbonyl C had the highest scores along PC1, O-alkyl C had the lowest, and OC, C/N ratio, TN, and TP were close to the 0 scale (Fig. 9b). Along PC2, C/N ratio and OC content showed the lowest scores, TN and TP contents showed the highest, and TK and C functional groups were close to the 0 scale. Since the proportion of PC1 was larger than that of PC2, the results of the factor loadings indicated that, although the chemical composition also influenced the decomposition of organic materials, the organic C functional groups played more important roles in their decomposition processes. Furthermore, the factor loadings of the parameters also suggested that the decomposition of woody peat (wP) was mainly affected by alkyl C and A/O-A ratio, while the decomposition of the mixture of woody peat and organic

manure (PM) was primarily influenced by aromatic C, aromaticity, and carbonyl C. In addition to alkyl C and A/O-A ratio, the decomposition of the mixture of woody peat and corn straw (PS) was also influenced by OC and C/N ratio. The decomposition of organic manure, corn straw, and their mixture (MS), on the other hand, was mainly affected by O-alkyl C, TN, TP, and TK.

#### *Residue-mixing effects on weight loss, OC, nutrients, and organic C functional groups of organic materials*

The SPI values for  $W_t$ , OC, TN, C/N ratio, TK, and TP during the incubation period were deviated from 0 (Fig. 10), suggesting non-additive effects in all the mixture treatments (PS, PM, and MS). Both PS and PM showed positive (synergistic) effects on  $W_t$  and OC, suggesting that blending

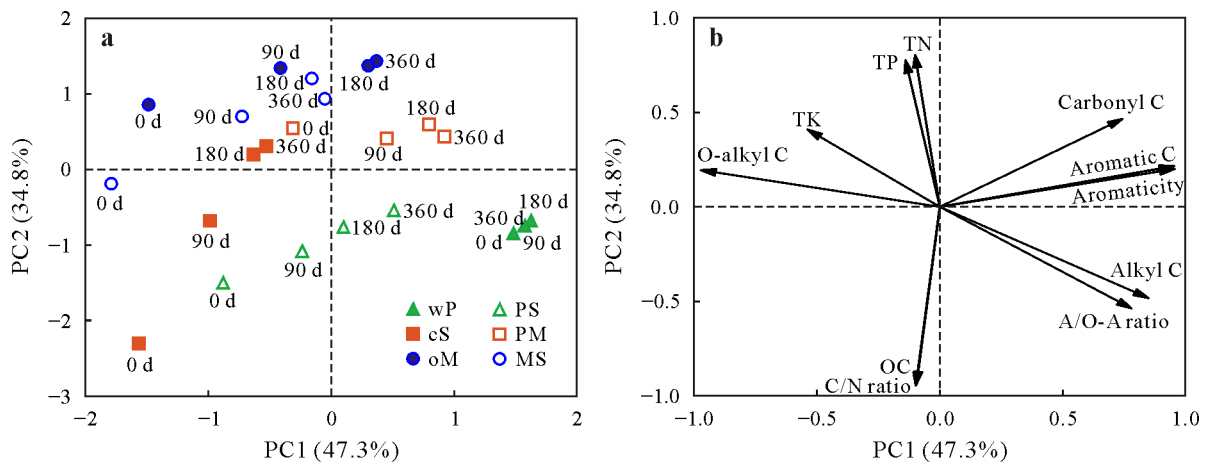


Fig. 9 Scatter plot (a) and factor loadings (b) of principal component (PC) analysis of chemical composition and organic C functional groups of organic materials (OM) in the different decomposition treatments with OM-containing litterbags buried in a newly reclaimed saline-sodic soil and incubated at 25 °C for 360 d. wP = woody peat; cS = corn straw; oM = organic manure; PS = woody peat + corn straw; PM = woody peat + organic manure; MS = organic manure + corn straw; TK = total K; TP = total P; TN = total N; A/O-A = alkyl C/O-alkyl C; OC = organic C.

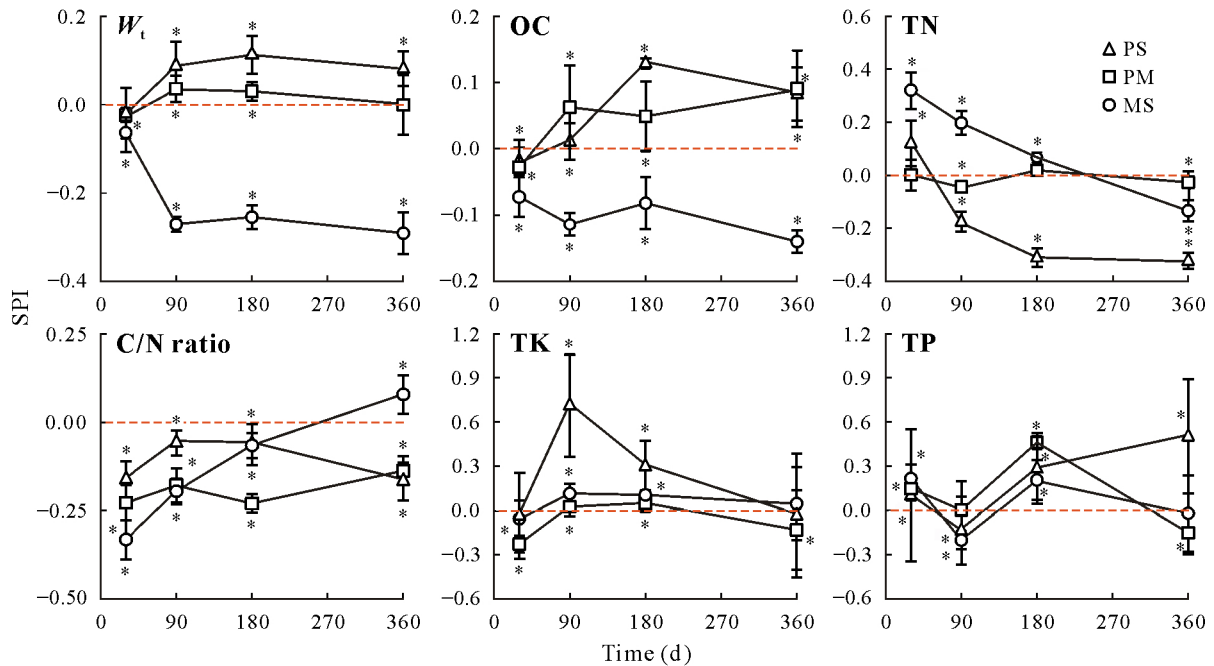


Fig. 10 Residue-mixing effects, as measured by strength of possible interactions (SPI), on the levels of weight loss percentage ( $W_t$ ), organic C (OC) content, total N (TN) content, C/N ratio, total K (TK) content, and total P (TP) content of organic materials (OM) in the decomposition treatments with OM-containing litterbags buried in a newly reclaimed saline-sodic soil and incubated at 25 °C for 360 d. Error bars are standard deviations of the means ( $n = 3$ ). The asterisk \* indicates a significant difference from 0 d according to one-sample  $t$ -test at  $P < 0.05$ . PS = woody peat + corn straw; PM = woody peat + organic manure; MS = organic manure + corn straw.

woody peat with corn straw or organic manure promoted OC accumulation and retarded weight loss. Conversely, MS exhibited negative (antagonistic) effects on OC and  $W_t$ , which indicated that mixing corn straw and organic manure accelerated the degradation of the mixture. Different from  $W_t$  and OC, PS only showed a synergistic effect on TN before day 30, and PM presented an antagonistic effect at days 90 and 360. In contrast, MS exhibited a significant synergistic effect on TN before day 180. All the mixture treatments showed an antagonistic effect on the C/N ratio

during the incubation, except MS on day 360. These results suggested that the addition of woody peat to corn straw and organic manure restrained the increase of TN in the mixtures, while mixing corn straw and organic manure promoted the accumulation of N. Meanwhile, a synergistic effect was predominant for TK and TP in PS, PM, and MS during the incubation period.

The SPI values for alkyl C, O-alkyl C, aromatic C, carbonyl C, A/O-A ratio, and aromaticity were also deviated from 0, indicating non-additive effects on the C functional

groups in the three mixture treatments (Fig. 11). For PS, a synergistic effect was predominant for alkyl C, aromatic C, carbonyl C, A/O-A ratio, and aromaticity, while an antagonistic effect was predominant for O-alkyl C. The synergistic and antagonistic effects in PS showed decreasing trends with incubation time. Although a synergistic effect was also observed for aromatic C, carbonyl C, A/O-A ratio, and aromaticity in PM, an antagonistic effect for alkyl C and a synergistic effect for O-alkyl C were predominant in this treatment, which differed from PS. Besides, the synergistic effect was much weaker in PM compared to PS. Similar to PS, a synergistic effect was predominant for alkyl C, aromatic C, carbonyl C, A/O-A ratio, and aromaticity, and an antagonistic effect was predominant for O-alkyl C in MS.

## DISCUSSION

### *Decisive factors in the decomposition of organic materials*

Application of exogenous organic materials is an important and efficient agricultural practice to improve SOM content and soil fertility (Yue *et al.*, 2023). However, due to significant differences in quality, chemical composition, and molecular structure, different organic materials exhibit markedly different decomposition rates (Chen *et al.*, 2015; Finn *et al.*, 2015), thus possessing different abilities to improve SOM content and soil productivity. To evaluate the efficacy of various organic materials in enhancing soil quality on newly reclaimed lands, it is beneficial to investigate their

primary recalcitrance or vulnerability to decomposition. In this study, corn straw, organic manure, and woody peat showed distinct decomposition rates and residence times in the saline-sodic soil (Fig. 1). Organic material weight loss percentage was significantly positively correlated with its TN content but significantly negatively correlated with its C/N ratio (Fig. 7). That is, the higher its TN content or the lower its C/N ratio, the faster the decomposition rate of an organic material. This result was consistent with the finding of Berg and McClaugherty (2020). Furthermore, the organic material decomposition rate was also significantly influenced by the composition of organic functional groups. The higher the contents of alkyl C and aromatic C, the slower the decomposition rate, whereas the higher the content of O-alkyl C, the faster the decomposition rate (Fig. 8). These results are consistent with traditional viewpoints (Finn *et al.*, 2015; Barré *et al.*, 2016), and it can be deduced that alkyl C and aromatic C are recalcitrant organic functional groups to a certain extent, while O-alkyl C is a labile organic functional group. Besides, the decomposition of organic materials, to some extent, was also influenced by the TK and TP contents (Figs. 7 and 8). However, the PCA analysis clearly indicated that compared to chemical composition and elemental contents, C functional groups played a more important role in the decomposition of organic materials (Fig. 9). The organic materials with higher relative abundances of aromatic C, alkyl C, and carbonyl C, as well as higher aromaticity and A/O-A ratio, tended to decompose more slowly.

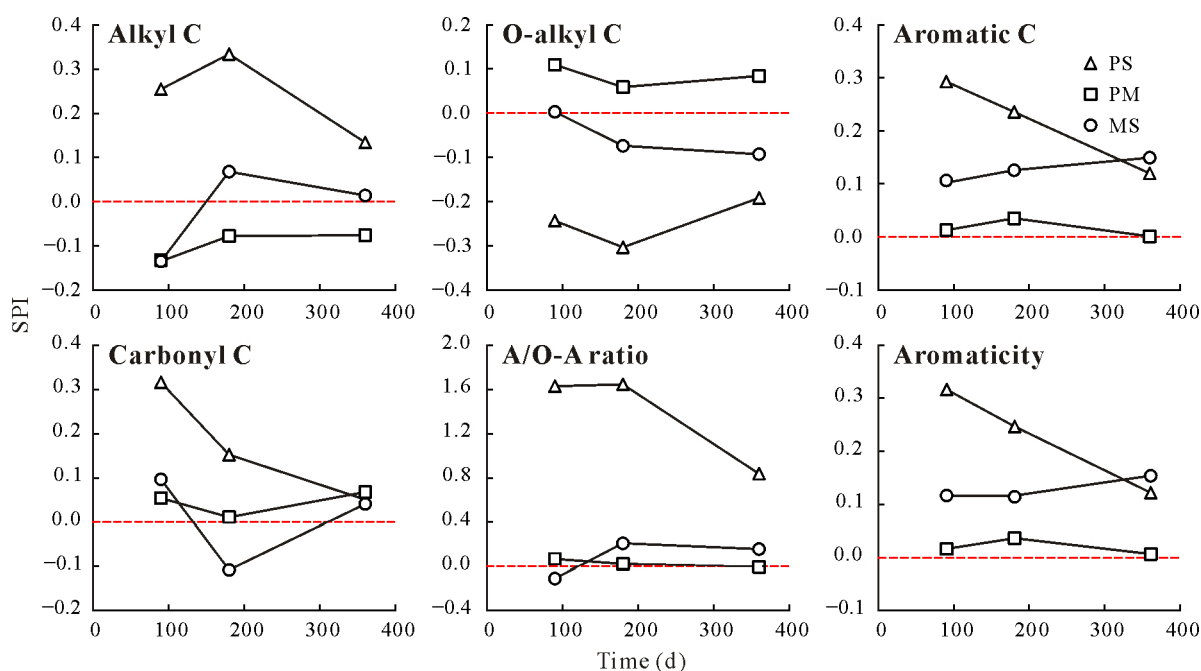


Fig. 11 Residue-mixing effects, as measured by strength of possible interactions (SPI), on the relative abundances of alkyl C, O-alkyl C, aromatic C, and carbonyl C, alkyl C/O-alkyl C (A/O-A) ratio, and aromaticity of organic materials (OM) in the mixed decomposition treatments with OM-containing litterbags buried in a newly reclaimed saline-sodic soil and incubated at 25 °C for 360 d. PS = woody peat + corn straw; PM = woody peat + organic manure; MS = organic manure + corn straw.

Due to the higher relative abundance of labile O-alkyl C (Figs. 4 and 5), corn straw exhibited a faster decomposition rate than organic manure and woody peat (Fig. 1), even though its C/N ratio was the highest (Table SIII). Organic manure also contained a high relative abundance of labile O-alkyl C (Figs. 4 and 5), was rich in nutrients of N, P, and K, and had a low C/N ratio (Table SIII), thereby displaying a rapid decomposition rate and losing more than 45% of its OC in the saline-alkali soil after the one-year incubation (Figs. 1 and 2). Compared to corn straw and organic manure, woody peat was rich in alkyl C and aromatic C, with a much lower relative abundance of O-alkyl C (Fig. 4), which resulted in the slowest decomposition rate in the saline-alkali soil (Fig. 1). After one year of decomposition, it only lost 6.4% of its OC (Figs. 1 and 2), demonstrating a higher capacity to enhance the content of passive SOM.

Although the CP NMR technique is thought to be not reliably quantitative for complex aromatic structures (Mao and Schmidt-Rohr, 2004), it is sufficient to assess the influences of C functional groups on the decomposition capacity of different organic materials. Even if the CP NMR technique may underestimate the aromatic C (Gerke, 2018), it is unlikely that the aromatic C measured in corn straw and organic manure would exceed O-alkyl C in relative abundance. Many studies also suggested that fresh crop straw and organic manure mainly consist of labile O-alkyl C (Zhou *et al.*, 2015; Xu *et al.*, 2017). For woody peat, the recalcitrant aromatic C determined by the CP NMR technique exceeded 26% (Figs. 4 and 5). The more precise and quantitative direct polarization (DP) NMR technology would only serve to further confirm (and likely increase) the abundance of the aromatic C (Gerke, 2018). Consequently, the use of CP NMR technique instead of DP NMR ought to have no significant influence on the above conclusions.

#### *Non-additive effects of mixed organic materials in newly reclaimed soils*

Low SOM content is a main obstacle restricting the efficient utilization of newly reclaimed lands (Chen Y P *et al.*, 2019). Increasing the SOM content through the application of abundant organic materials is one of the key practices to improve the soil quality of these lands (Zhang, 2023). However, how to efficiently and durably sequester more C from organic materials in the newly reclaimed soils is a topic of great concern (Sierra *et al.*, 2024). According to previous discussion, different organic materials with varied primary recalcitrance and chemical composition have distinct decomposition rates. Therefore, to facilitate rapid and sustained increases in SOM content and soil fertility, application of recalcitrant organic materials, such as woody peat, is advisable (Fu *et al.*, 2021; Lin *et al.*, 2022). Nevertheless, the lower decomposition rate and OC loss rate of

woody peat also imply that it may be challenging to supply an adequate amount of active organic matter to the soil. Clearly, the combined application of recalcitrant woody peat with easily decomposable corn straw or organic manure can mitigate their drawbacks, since their mixtures have the potential to simultaneously improve active and passive SOM contents (Zhao *et al.*, 2020). The present study showed that the combination of woody peat with corn straw or organic manure led to a more balanced ratio of active to passive organic matter (Figs. 4 and 5), significant reduction in OC loss (Table I), and improvements in TN (Fig. 2), aromaticity and A/O-A ratio (Fig. 6), and C/N ratio (Table I). This is also the reason why many studies have found that, compared to the sole use of woody peat, the combined application of woody peat with labile organic materials such as straw and organic manure led to higher soil fertility and crop yields (Zhao *et al.*, 2020; Kang *et al.*, 2021; Li *et al.*, 2021; Zhang *et al.*, 2021; Duan *et al.*, 2023).

However, this result may be attributed not only to the average mixing effect or additive effect but also to the non-additive effect. An additive effect indicates that there is no interaction between the constituent residues during the decomposition, while a non-additive effect implies that interactions occur between the residual components, which can either promote or inhibit the decomposition of organic materials (Zhou *et al.*, 2019). In this study, significant non-additive effects (either synergistic or antagonistic) were observed on the decomposition rate, chemical composition, and organic C functional groups when woody peat, corn straw, and organic manure were mixed pairwise (Figs. 10 and 11), suggesting that intensive interactions exist between these organic materials. Due to the interaction between woody peat and corn straw, the mineralization of labile organic components (O-alkyl C) and the release of N from the woody peat-corn straw mixture were promoted, as well as the accumulation of recalcitrant organic components (alkyl C and aromatic C). Gerke (2018) argued that humic substances are synthesis products of phenolic substances with an aromatic core; therefore, they possess a unique ability to incorporate organic and inorganic molecules derived from plant residues to form stable complexes. Woody peat contains a large amount of humus (Zheng *et al.*, 2018) and is characterized by abundant of aromatic C (Figs. 4 and 5) and high aromaticity (Fig. 6). Hence, the combination of woody peat with corn straw could promote the absorption of degradation products from corn straw by woody peat, retard the decomposition rate of the mixture, and facilitate the accumulation of organic matter in the soil. Piccolo (2002) proposed that humic substance is a supramolecular assembly of small molecules originating from various biological residues and linked together mainly through hydrophobic interactions. The degree of polymerization of humic substance has a crucial impact

on its recalcitrance (Piccolo *et al.*, 2018). It has also been demonstrated that small molecules incorporated into the most stable SOM fractions were primary aliphatic compounds (Almendros *et al.*, 1998; Piccolo, 2002). Thus, the elevated relative abundance and signal intensity in the NMR spectrum of alkyl C in PS (Figs. 4 and 5) were presumptively attributable to the sorption of aliphatic compounds by woody peat. Moreover, the integration of hydrophobic components played a crucial role in impeding the rapid microbial mineralization of the organic materials, thereby enhancing their longevity within the soil (Piccolo, 2002). Due to its high content of hydrophobic compounds, such as polycyclic compounds and aliphatic hydrocarbons, peat typically exhibits pronounced hydrophobicity (Wu *et al.*, 2020). Hence, the combination of woody peat and corn straw might augment the resistance of the mixture to microbial degradation (Fig. 1). As a result, the mixing of woody peat with corn straw significantly decelerated the decomposition rate, extended the residue time in the soil, and intensified the humification degree (Figs. 1 and 6).

Compared to corn straw, the mixture of woody peat and organic manure also presented a clear non-additive effect (Figs. 10 and 11), mitigated the decomposition of the mixture and the release of OC (Figs. 1 and 2), and thereby prolonged their residence time in the soil. The reason for this result may lie in the fact that organic manure is a product of a certain period of humification processes, containing humic substances (Antil *et al.*, 2013), and compared to corn straw, it contained more alkyl C and aromatic C (Figs. 4 and 5). Therefore, according to the theory of Gerke (2018), the aromatic woody peat could absorb more alkyl C and aromatic C from organic manure as compared to corn straw. This was supported by the results shown in Figs. 4 and 5 that PM showed stronger intensities in alkyl C and aromatic C than PS. However, the non-additive effects of woody peat and organic manure were significantly weaker than those of the woody peat and corn straw (Figs. 10 and 11). The reason for this result is probably multifaceted. Firstly, organic manure contained less labile components and more recalcitrant components compared to corn straw (Figs. 4 and 5), resulting in a much slower decomposition rate (Fig. 1). Secondly, organic manure typically has a higher hydrophilic carbonyl C content than straw, making it difficult for the adsorbed small molecules to be protected from decomposition through hydrophobic interactions (Piccolo, 2002). Additionally, N may also play a role in this outcome, because TN was negatively correlated with alkyl C and aromatic C but positively correlated with O-alkyl C (Fig. 8). Studies have shown that under high N conditions, synergistic effects can shift to additive or antagonistic effects, thereby slowing down the decomposition of mixed organic materials (Wu *et al.*, 2023).

Different from the woody peat mixtures, the combination of corn straw and organic manure significantly accelerated

the decomposition of organic materials, reduced the residual time, and exhibited significant antagonistic effects (Figs. 1 and 11). This outcome can be partially attributed to the high relative abundance of labile O-alkyl C (Fig. 5) and the reduced levels of aromaticity and humification (A/O-A ratio) (Fig. 6). Generally, recalcitrant organic matter is characterized by high aromaticity and A/O-A ratio (Kögel-Knabner, 1997; Quideau *et al.*, 2000; Cerli *et al.*, 2008; Thomsen *et al.*, 2008; Zheng *et al.*, 2021). Moreover, according to Gerke (2018) and Piccolo (2002), the observed result may also be influenced by the lack of an aromatic core of humic substance and the absence of hydrophobic protection.

The findings of this study suggest that relying solely on corn straw or organic manure to improve the SOM content of newly reclaimed land would require a considerable quantity of organic materials and incur significant time costs. For instance, after continuous application of crop straw at a rate of 5 Mg ha<sup>-1</sup> year<sup>-1</sup> for 30 years, Luo *et al.* (2020) observed only a 7.1%–15.4% increase in the SOC content of a Typic Hapludoll. Xin *et al.* (2016) reported that after 23 years of continuous application of organic fertilizer, the SOC content increased slightly from 3.81 to 9.08 g kg<sup>-1</sup>. If these two materials are applied together, higher quantities and a longer time may be needed due to their antagonistic effect (Figs. 10 and 11). In contrast, this study demonstrated that co-application of woody peat and easily decomposable organic material, such as corn straw and organic manure, can promote the accumulation of recalcitrant components and slow down the decomposition of organic materials (Figs. 1 and 11). Consequently, to rapidly improve the organic matter and fertility levels of newly reclaimed arable soils, it is recommended to apply woody peat in combination with easily decomposable organic materials such as crop straw and organic fertilizers. However, this study was conducted using the litterbag incubation method under constant temperature conditions (25 °C), which may not fully reflect the real decomposition environment of the saline-sodic soil. Environmental factors, such as temperature, and the interaction of organic matter with clay minerals, can significantly affect decomposition dynamics (Baldock and Skjemstad, 2000; Karhu *et al.*, 2010). Therefore, further research is essential to investigate the decomposition of woody peat and its mixture with other organic materials under the real conditions in newly reclaimed arable lands, using more representative methods.

## CONCLUSIONS

Woody peat, corn straw, and organic manure showed different decomposition rates in the saline-sodic soil, which were primarily influenced by their organic C functional groups rather than chemical compositions. Due to the highest relative abundances of recalcitrant alkyl C and aromatic C,

woody peat exhibited the slowest decomposition rate. In contrast, because of the high relative abundance of labile O-alkyl C, corn straw and organic manure showed fast decomposition. Compared to the incorporation of woody peat, corn straw, and organic manure alone, the co-application of woody peat and labile organic materials was more effective improving SOM content and its composition, TN, aromaticity, and the A/O-A and C/N ratios, and reducing OC loss. This result was mainly attributed to the non-additive effect, which significantly promoted the accumulation of recalcitrant compounds, slowed down the decomposition of organic materials, and extended the retention of OC in soil. Therefore, to rapidly improve the organic matter and fertility levels of newly reclaimed arable soils, it is recommended to apply woody peat in combination with easily decomposable organic materials such as corn straw and organic manure.

#### DECLARATION OF COMPETING INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### SUPPLEMENTARY MATERIAL

Supplementary material for this article can be found in the online version.

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