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Organic fertilizer reduced carbon and nitrogen in runoff and buffered soil acidification in tea plantations: Evidence in nutrient contents and isotope fractionations

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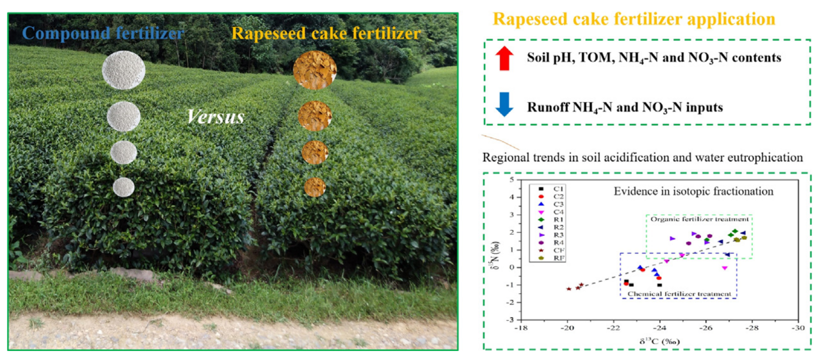
HIGHLIGHTS

- Organic fertilizer effectively preserved soil C and N pools of the tea plantation.
- Organic fertilizer substantially reduced the NH_4-N and NO_3-N inputs in the tea plantation runoff.
- Organic fertilizer and compound fertilizer treatments both had strong impacts on the $\delta^{13}C$ and $\delta^{15}N$ of the tea plantation soil.

GRAPHICAL ABSTRACT



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ABSTRACT

Carbon (C) and nitrogen (N) inputs to farmland via fertilizer application are potential sources of C and N that influence soil acidification and water eutrophication. A pilot study was conducted to compare the effects of compound fertilizer and rapeseed cake organic fertilizer on C and N preservation in the soils and runoff of a tea plantation as well as the C and N isotopic fractionation in soils over the three annual cycles of fertilization and tea-leaf harvest. Overall, rapeseed cake organic fertilization effectively increased the pH, total organic matter, NH_4-N and NO_3-N in soils by 2.19–4.29%, 8.04–21.14%, 53.65–100.32% and 5.74–54.08%, respectively, but decreased NH_4-N inputs in runoff by 10.36–25.12% and NO_3-N inputs in runoff by 8.94–24.10% relative to the same rate of pure N in compound fertilizer. Before fertilization in February, the average $\delta^{13}C$ and $\delta^{15}N$ were −25.15‰ and 1.88‰, while after a full year of fertilization and tea-leaf harvesting in October, the average soil $\delta^{13}C$ and $\delta^{15}N$ contents were −23.83‰ and −0.33‰ after compound fertilization and −26.22‰ and 1.64‰ after rapeseed cake organic fertilization, respectively, indicating the evident effects of fertilization on the isotopic fractionation in soil. In addition, the fractionation extent was positively associated with the fertilization rates under both fertilizers. However, the two fertilization types had different effects on the C and N isotope fractionations, with rapeseed cake organic fertilization contributing more to $\delta^{13}C$ (21.07–81.80%) but less to $\delta^{15}N$ (18.20–78.93%) and compound fertilization presenting the opposite results (1.88–46.18% and 53.82–98.12%, re-

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spectively). This study demonstrates that rapeseed cake organic fertilization can better preserve soil C and N pools while reducing their runoff in tea plantations, which may greatly hinder the regional soil acidification and water eutrophication trends.

1. Introduction

Tea trees (*Camellia sinensis*) are a valuable cash crop that is widely planted in China. The total area of tea plantations in China reached more than 2.85 million ha and the yield exceeded 2.46 million tons in 2017, which made China the largest producer and exporter of tea in the world (NBSC, 2017). Tea plantations are widely distributed and require large fertilizer inputs for the promotion of tea leaf growth. The yield and quality of tea, one of the most popular beverages consumed worldwide, directly affect the income of hundreds of thousands of tea farmers (Liu et al., 2019). Unlike cereal crops, which are harvested for grain yield, tea plants are always plucked for young shoots and leaves and thus demand more N for high yield and quality components (Mudau et al., 2007; Ruan et al., 2010). As a result, high fertilizer application rates have generally been used to improve the output and quality of tea; however, increasing the usage of fertilizers does not always proportionally increase the tea yield. At the same time, this causes environment problems, such as soil acidification and nitrate pollution of the waterbody (Saraswathy et al., 2007; Ji et al., 2018). Field investigations of tea farms have shown that the nitrogen fertilizer application rates are 0–1200 kg N/ha/yr (average of 533 kg N/ha/yr) in Chinese tea plantations, which far surpasses the amounts required (Wu et al., 2016). In Japanese tea plantations, chemical fertilizers are also being applied at far greater rates than the actual annual demands of mature tea trees at more than 200 kg N/ha/yr (Tokuda and Hayatsu, 2000). Moreover, similar overfertilization of tea plantations occurs throughout the world. Residual C and N from fertilizer can leach into runoff water and substantially contribute to the emission of pollutants into surrounding water bodies and soils (Smith et al., 2007; Li et al., 2016; Yang et al., 2018), resulting in serious agricultural nonpoint pollution (Oh et al., 2006; Sun et al., 2012; Wang et al., 2020). Therefore, effective management practices must be implemented to reduce excessive fertilizer inputs.

Many studies have documented fertilizer use in tea plantations and investigated alternative measures for reducing the amount of fertilizer applied. Han et al. found that three slow-release fertilizers coupled with a nitrification inhibitor could improve N use efficiency and simultaneously reduce environmental pollution (Han et al., 2008). In Sri Lanka, Tennakoon et al. formulated dual plant growth-promoting rhizobacteria inoculants to enhance the growth and yield of tea without the need for large fertilizer inputs (Tennakoon et al., 2019). Similar observations have been reported in which organic fertilizer had a lower nitrification potential than inorganic nitrogen fertilizer at the same nitrogen level because the nutrient content in the organic fertilizer was lower than that in the pure chemical fertilizer (Heeb et al., 2006; Klop et al., 2012; Liang et al., 2012). Our previous work found that proper management via the substitution of ammonium fertilizer for organic fertilizer could effectively mitigate soil acidification and nutrient deficiency in a green tea plantation in Zhejiang Province of China to promote tea yield and quality (Xie et al., 2019). The study also showed that the detrimental environmental impacts of tea cultivation could be effectively reduced by appropriate management with decreasing fertilizer application. However, more data are still needed to assess the intrinsic effects of fertilizer. Compared with conventional soil properties, stable isotope ratios can reflect differences in the growth conditions of plants and remain stable in samples before and after processing (Pilgrim et al., 2010).

Carbon (C) and nitrogen (N) stable isotope ratios ($\delta^{13}C$ and $\delta^{15}N$) are widely used as useful tools for estimating the source and composition of soil C and N in various ecosystems. A large number of studies have shown that $\delta^{13}C$ and $\delta^{15}N$ have advantages in studying the origins of cash crops, nutrient conversion and water use efficiency. For example, Choi et al. conducted a study on loblolly pine fertilization and irrigation at the Southeast Tree Research and Education Site in the USA and concluded that irrigation and fertilization affected the $\delta^{13}C$ and $\delta^{15}N$ values in the plant and soil samples (Choi et al., 2005). Spangenberg et al. traced changes in soil water availability in vineyards by using the $\delta^{13}C$ and $\delta^{15}N$ compositions of dried wines (Spangenberg and Zufferey, 2018). Ni et al. used isotopic signatures to determine the geographical origins of green tea collected from different areas in China, and the prediction accuracies of all methods were greater than 70% (Ni et al., 2018). In tea plantation soil ecosystems, the application of chemical fertilizer not only directly changes the carbon (C) and nitrogen (N) inputs but also further influences the C and N isotopic compositions ($\delta^{13}C$ and $\delta^{15}N$) of soils. Quantitative assessments of soil constituents are widely used and have been enhanced by stable isotopic techniques (Senbayram et al., 2008; Gattinger et al., 2012). The application of $\delta^{13}C$ and $\delta^{15}N$ in tracing C and N sources is mainly based on the distinct isotopic composition signatures relative to potential external input sources. Collins et al. (1999) found that the $\delta^{13}C$ values in moldboard-tilled soil were higher than those in uncultivated soil due to isotopic fractionation of soil organic carbon. Awiti et al. (2008) observed high and low $\delta^{13}C$ and $\delta^{15}N$ values in low- and high-fertility soil classes, respectively. Similar results were found by Busari et al., who reported that conservation tillage practices, such as no tillage and integrated application of organic and inorganic fertilizers, were good strategies for reducing soil C and N emissions, as proven by C and N isotopic sequestration in soils (Busari et al., 2016). The substitution of fertilizer types can potentially change the general inputs of C and N to soils, and a study found that in an organic microaggregate treatment, the organic carbon storage of soil microaggregates increased by 39.8% compared with the no fertilizer treatment (Huang et al., 2010). Therefore, carbon sequestration in farmland soils can be regulated by organic fertilizer substitution measures, which have great potential for carbon sequestration in global ecosystems (Wiesmeier et al., 2015; Li and Han, 2016; Di et al., 2020).

As one of the top three non-alcoholic drinks worldwide and the major cash crop in a number of developing countries, such as China, India, Sri Lanka, and Kenya, tea has shown increased consumption worldwide world due to the increasing interest in its health benefits (Su et al., 2015; Yan et al., 2020). However, in tea-producing countries, environmental problems associated with tea plantations, such as soil acidification and nitrogen leaching, are becoming apparent. High rates of chemical fertilizer application are likely to cause low fertilizer use efficiency and environmental pollution (Wang et al., 2010; Fu et al., 2012; Chen et al., 2017). The majority of previous studies mainly focused on soil nutrient concentration emissions caused by chemical fertilizer application, whereas few quantitative and accurate evaluations have been performed on the C and N change characteristics in tea plantation soil and runoff. Moreover, few studies have explored the dynamic variations of C and N and associated driving mechanisms under different fertilization rates. Tea plantations are usually fertilized multiple times throughout the year, which is consistent with the breeding-harvest cycles. The inputs and outputs of nutrients are obvious and easily observed, and there are few other interfering factors in tea plantations. As a result, soil and runoff water can quickly respond to the C and N changes resulting from fertilization and sample collection is easier to control from tea plantations compared with other types of farmland, such as rice paddies. Therefore, we hypothesized that 1) organic fertilization is a more effective way to reduce C and N in runoff and buffer soil acidification during tea cultivation compared with traditional compound fertilization in tea plantations; and 2) the variation characteristics of C and N under fertilization treatment in tea plantation soils could be well recorded and quantitatively evaluated by $\delta^{13}C$ and $\delta^{15}N$ isotopic techniques. The objectives of this study were to examine the above two hypotheses, and the results are expected to provide insights into how the rate and type of fertilization affects C and N preservation in soils and inputs to runoff water in tea plantations.

2. Materials and methods

2.1. Experimental site and design

The experimental site used for this study is a typical green tea plantation located in the suburbs of Shaoxing City, Zhejiang Province, China (Fig. 1). The site has an average altitude of less than 170 m, average slope of less than 5°, annual temperature of 15 °C and average precipitation of 1200 mm, making it suitable for tea-tree growing. Soils in this area are mostly yellow brown, and tea plantations have been cultivated in the past twenty years. As a typical subtropical humid monsoon climate of this area, spring is warm and humid and summer is hot. Heavy rainfall occurs frequently but in a short period of time during summer (Fig. 2). Pilot experimental tea plots (3 m W*7 m L each) were selected in areas where tea trees have grown for more than ten years and have not been fertilized for five years, thus guaranteeing the reliability of the study in terms of eliminating the influence of external factors. Compound fertilizer and rapeseed cake organic fertilizer were

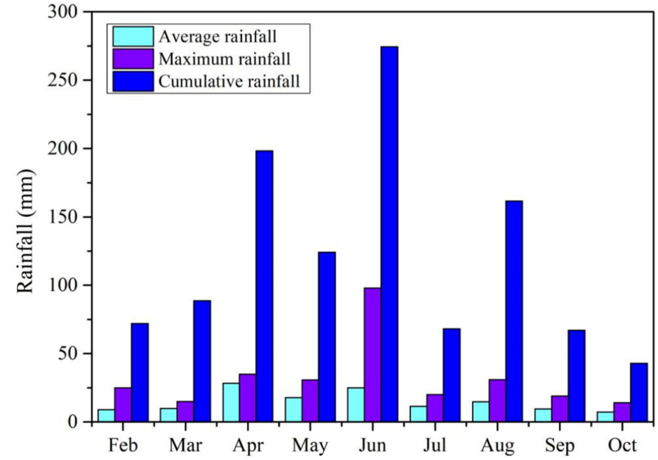

Fig. 2. Average rainfall, maximum rainfall, and cumulative rainfall during the experiment in the study area.

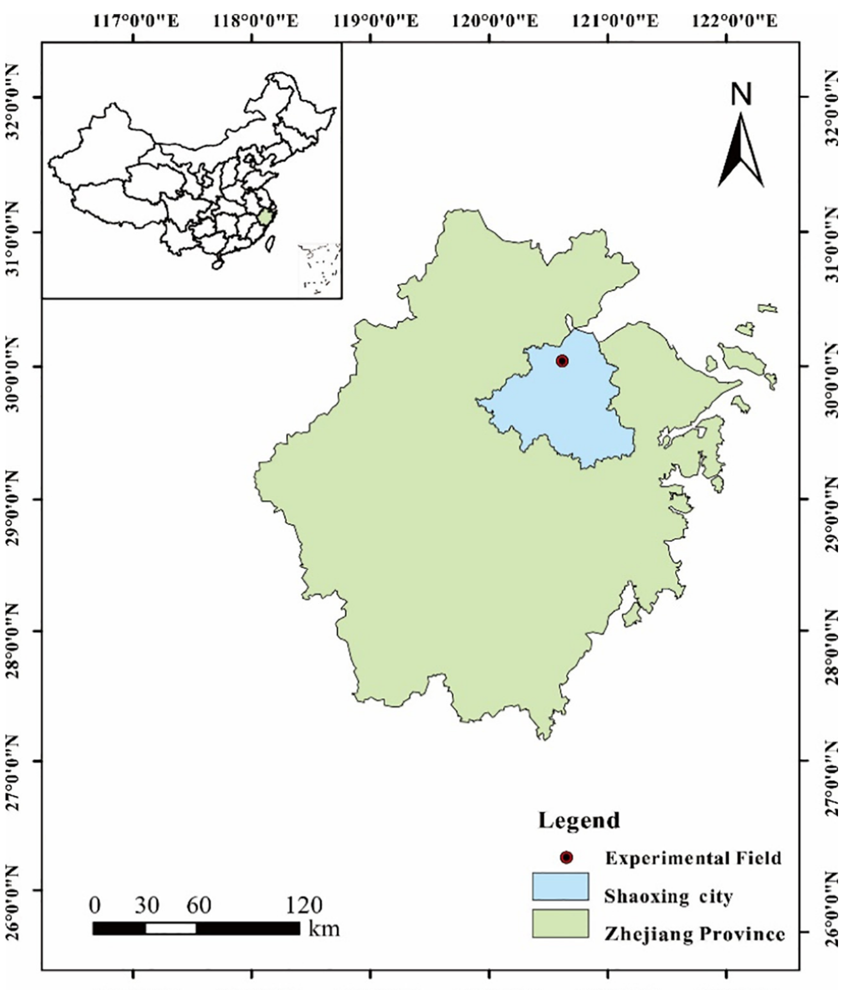

Fig. 1. Location of the experimental site.

added to the plots at four rates (levels). The baseline doses of compound fertilizer and rapeseed cake organic fertilizer were used as the 100% treatment groups (named C1 and R1, respectively), while 80% (C2 and R2), 50% (C3 and R3) and 20% (C4 and R4) of the baseline were designated as the other treatment groups. The baselines of 100% compound and rapeseed cake organic fertilization were designed to be consistent with the annual pure nitrogen application rate of 450 kg/ha (compound fertilizer has a N content of 150 g/kg; rapeseed cake organic fertilizer has a N content of 52.7 g/kg). A total of eight treatments were included, each repeated 3 times (Table 1). All the experimental plots, which had an area of 21 m^2, were randomly arranged with their length parallel to the slope. Fertilization was performed at the end of March, May, and July; thus, the proportion of the year with fertilization was 5:3:2. All fertilizers were added using artificial broadcasting in ditches at a depth of 15 cm between two rows of tea plants. Ten liter runoff buckets were placed at the bottom of each experimental plot across the direction of flow to collect the runoff water. Additionally, the volumes of rainfall collected in the buckets were recorded in detail at each rainfall event throughout the experiment from February to October of the year.

2.2. Sample collection and statistical analysis

Soil samples (0–20 cm) were collected at the end of February, April, June, August and October of 2017. Five subsamples with a "S" shape distribution in each plot were collected and then combined to form a composite sample. Soil samples were then air-dried, sieved, and ground using an agate mortar and pestle. Runoff water samples were collected after each rainfall event, stored in a refrigerator at 4 °C and transported to the laboratory for testing within five days. Soil pH was measured using a 1:2.5 ratio of soil to deionized water; soil total organic matter (TOM) was measured using an elemental analyzer; NH_4-N and NO_3-N in 1M KCl soil extracting solution, as well as in filtered runoff water were analyzed using an automatic chemical analyzer (Tian, 1997). The C and N contents and $\delta^{13}C$ and $\delta^{15}N$ levels of the soils were measured using a ThermoFinnigan MAT 253 instrument. The $\delta^{13}C$ and $\delta^{15}N$ was expressed as the differences between the isotopic ratios of a sample to the standard materials.

$$\delta^{13}C_{sample} = \{(^{13}C/^{12}C_{sample})/(^{13}C/^{12}C_{standard}) - 1\} \times 1000$$

$$\delta^{15}N_{sample} = \{(^{15}N/^{14}N_{sample})/(^{15}N/^{14}N_{standard}) - 1\} \times 1000$$

where $^{13}C/^{12}C_{sample}$, $^{13}C/^{12}C_{standard}$, $^{15}N/^{14}N_{sample}$ and $^{15}N/^{14}N_{standard}$ are the C and N isotope ratios in samples and standards, respectively. The standard substance for C and N isotope determination was Vienna PDB and atmospheric N_2, each with 0‰ in values of $\delta^{13}C$ and $\delta^{15}N$. All samples collected from the experimental plots were analyzed, each with three repetitions to eliminate the effect of potential uncertain influencing factors on $\delta^{13}C$ and $\delta^{15}N$. Statistical analyses were carried out using SPSS V20, and correlation analyses between the rainfall and runoff water NH_4-N and NO_3-N contents were assessed by Pearson's correlation analysis ($P \leq 0.05$).

Table 1
Fertilizer types and application amounts in the fertilizer treatments.

Treatment name	Fertilizer treatment	Amount of fertilizer applied (kg/ha)
C1	100% compound fertilizer	3000
C2	80% compound fertilizer	2400
C3	50% compound fertilizer	1500
C4	20% compound fertilizer	600
R1	100% rapeseed cake fertilizer	8539
R2	80% rapeseed cake fertilizer	6831
R3	50% rapeseed cake fertilizer	4270
R4	20% rapeseed cake fertilizer	1708

Note: "C" and "R" represent compound and rapeseed cake fertilizer, respectively; and the baselines of 100% compound and rapeseed cake organic fertilization were designed to be consistent with the annual pure nitrogen application rate of 450 kg/ha.

. Results

. Changes of soil pH, TOM, NH_4-N and NO_3-N contents

At the beginning of the experiment in February, fertilizer was not applied, and the average soil pH was 4.30 while the average TOM, NH_4-N and NO_3-N contents were 15.93 g/kg, 6.65 mg/kg, and 6.20 mg/kg, respectively (Fig. 3). No significant differences in any soil properties were observed between the plots, indicating homogeneous initial soil conditions. In October after a whole year of fertilization, the pH decreased to 4.12 while the TOM, NH_4-N and NO_3-N contents increased significantly to average values of 20.19 g/kg, 30.34 mg/kg and 19.02 mg/kg, respectively. The pH decreased by 0.70–7.60%, while TOM, NH_4-N and NO_3-N increased by 15.26–43.92%, 144–831% and 101–335%, respectively.

In general, the average pH values in the rapeseed cake organic fertilization plots were slightly higher than those of the compound fertilization plots at the same N addition rates (Table 1, Fig. 3). With reduced rates of compound and rapeseed cake organic fertilization, the decrease in pH due to fertilization was reduced, with significantly smaller decreases observed for the rapeseed cake organic fertilization plots than the compound fertilization plots. Compared to February, the average pH decrease after the last fertilization in October were 2.28%, 3.32%, 3.59% and 0.70% in R1, R2, R3 and R4, respectively, and 6.89%, 7.60%, 3.59% and 4.85% in C1, C2, C3 and C4, respectively. Fertilization increased the TOM contents of the soils, which was more pronounced in the organic fertilization plots, e.g., the TOM contents after the last fertilization were 23.10 g/kg, 22.56 g/kg, 20.88 g/kg and 19.46 g/kg in R1, R2, R3 and R4, respectively, and 19.91 g/kg, 18.62 g/kg, 18.96 g/kg and 18.02 g/kg in C1, C2, C3 and C4, respectively, which were higher than those at the beginning of the experiment without fertilization in February. The soil NH_4-N contents in the C1, C2, C3 and C4 treatments in October increased by 508%, 225%, 144% and 147%, respectively, compared with the same plots in February, while those in the R1, R2, R3 and R4 treatments in October increased by 831%, 395%, 353% and 248%. Similarly, the soil NO_3-N contents increased by 282%, 218%, 113% and 101% in the C1, C2, C3 and C4 treatments, respectively, and 335%, 256%, 226% and 123% in the R1, R2, R3 and R4 treatments, respectively. The NH_4-N and NO_3-N contents of the soils were simultaneously decreased with the reduced application rates of both fertilizers (Fig. 3).

. Changes of NH_4-N and NO_3-N inputs in runoff water

In general, the NH_4-N contents were significantly lower than the NO_3-N contents in runoff water in every cycle of fertilization (Fig. 4). Fertilization significantly increased the N inputs in runoff water after fertilization in April and August. Before fertilization, the contents of NH_4-N and NO_3-N in the runoff water in February were 0.78 mg/L and 1.24 mg/L, respectively, while after the last fertilization in October, the contents of NH_4-N and NO_3-N in runoff increased to 1.11 mg/L and 2.55 mg/L, respectively (Fig. 4). After a whole year of fertilizer applications, the average contents of NH_4-N in the C1, C2, C3, and C4 treatments were 72.46%, 46.04%, 30.44% and 11.07%, respectively, while those in the R1, R2, R3 and R4 treatments were 60.85%, 32.44%, 36.77% and 53.84%, respectively, higher than those before fertilization. With reduced fertilization rates, the N inputs to runoff water decreased, e.g., with the average contents of NH_4-N in the C1, C2, C3, and C4 treatments at 1.30 mg/L, 1.20 mg/L, 1.04 mg/L and 0.83 mg/L, respectively, while those for the R1, R2, R3 and R4 treatments at 1.21 mg/L, 1.14 mg/L, 1.10 mg/L and 1.09 mg/L, respectively.

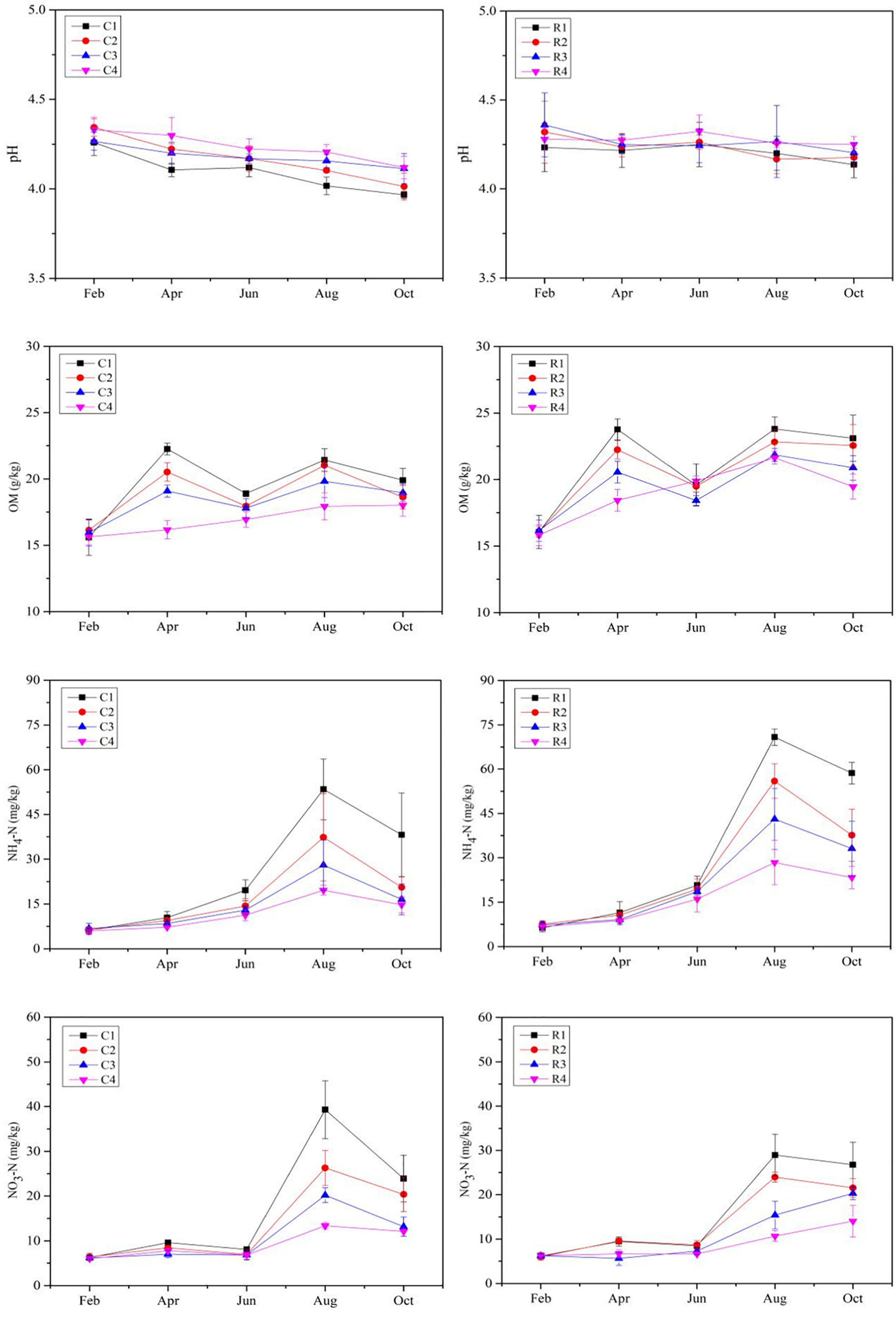

Fig. 3. pH and TOM, NH_4-N and NO_3-N contents in the soils of various experimental plots. Treatment details refer to Table 1.

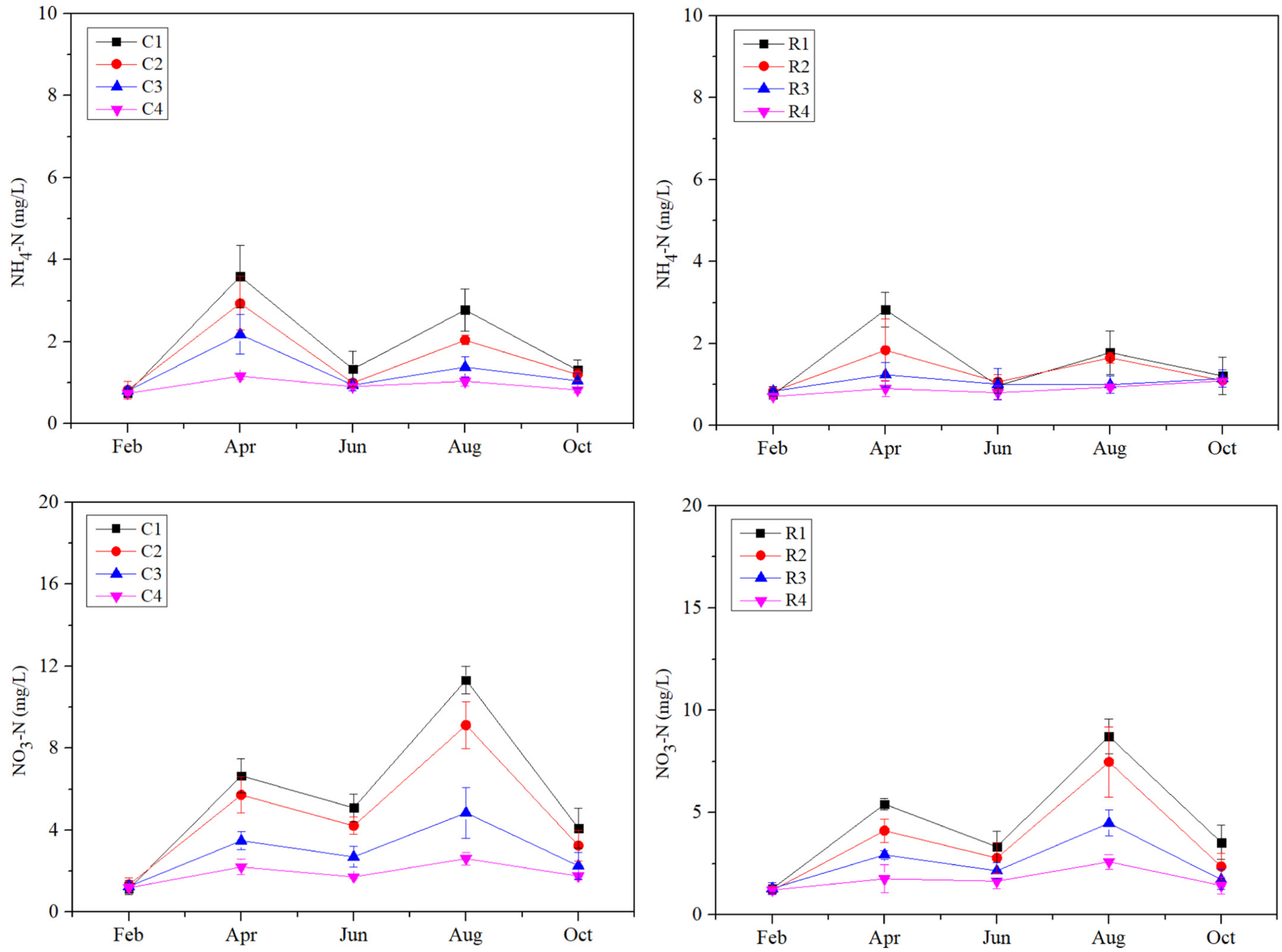

Fig. 4. NH_4-N and NO_3-N contents in the soils of various experimental plots. Treatment details refer to Table 1.

. Changes in the C and N contents and their isotopic compositions in soils

Before fertilization in February, the C contents of tea plantation soils ranged from 15.96–18.25 g/kg and averaged 16.71 g/kg and the N contents ranged from 0.75–1.28 g/kg and averaged 0.99 g/kg. After a full year of fertilization, the C contents of tea plantation soils changed to 18.77–20.72 g/kg and averaged 19.57 g/kg in the compound fertilizer treatments and 21.04–24.82 g/kg and averaged 23.02 g/kg in the rapeseed cake organic fertilizer treatments. The N contents ranged from 2.04–2.26 g/kg (average of 2.18 g/kg) in the compound fertilizer treatments and 2.07–2.31 g/kg (average of 2.17 g/kg) in the rapeseed cake organic fertilizer treatments (Table 3). After fertilization, the C contents in the rapeseed cake organic fertilizer treatments were significantly higher than that of the compound fertilizer treatments. However, there was no significant difference in the N contents between the rapeseed cake organic fertilizer treatment groups and the compound fertilizer treatment groups at the same N addition rates.

The application of fertilizer changed the δ^{13}C and δ^{15}N compositions of soil via C and N inputs from external sources, and the isotopic compositions of soil changed with the fertilization rates (Fig. 5). Before fertilization in February, the average δ^{13}C and δ^{15}N in soils of all treatment groups were relatively uniform, thus reflecting the homogeneity of the soil in the study area. The isotopic characteristics of C ranged from −25.45‰ to −24.40‰, with an average of −25.15‰, and those of N ranged from 1.29‰ to 2.37‰, with an average of 1.88‰. After a whole cycle of tea tree fertilization, the average δ^{13}C and δ^{15}N of the soil changed to −25.03‰ and 0.65‰. In the rapeseed cake organic fertilizer treatment groups, the δ^{13}C was significantly more negative while the δ^{15}N was significantly more positive compared with the compound fertilizer treatment groups, i.e., the average soil δ^{13}C and δ^{15}N of the rapeseed cake organic fertilizer treatment groups were −26.79‰ and 1.83‰ for R1, −27.08‰ and 1.39‰ for R2, −25.34‰ and 1.67‰ for R3, −25.69‰ and 1.65‰ for R4, respectively, while the average soil δ^{13}C and δ^{15}N of the compound fertilizer treatment groups after fertilization were −23.11‰ and −0.93‰ for C1, −23.26‰ and −0.56‰ for C2, −23.59‰ and −0.19‰ for C3, −25.39‰ and 0.36‰ for C4, respectively.

. Discussion

. Effects of fertilization types and rates on soil properties

This study was conducted to compare the effects of compound fertilizer and rapeseed cake organic fertilizer under different rates on environmental variables, including the soil properties, nutrient loss from soils to runoff water and the δ^{13}C and δ^{15}N of soil in a tea plantation. As a multiharvest agricultural product, tea trees require more nitrogen than most other crops, and fertilization applications occur several times before and after each tea harvest. A complete fertilization application cycle over a harvest year provided a thorough understanding of the environmental effects of compound fertilization and rapeseed cake organic fertilization. Tea plantation soil acidification is a serious problem worldwide and has been reported across a variety of ecosystems and regions (Kirk et al., 2010; Yan et al., 2018; Yang et al., 2018). Because tea is a unique, perennial and commercial crop that prefers acidic soil, tea

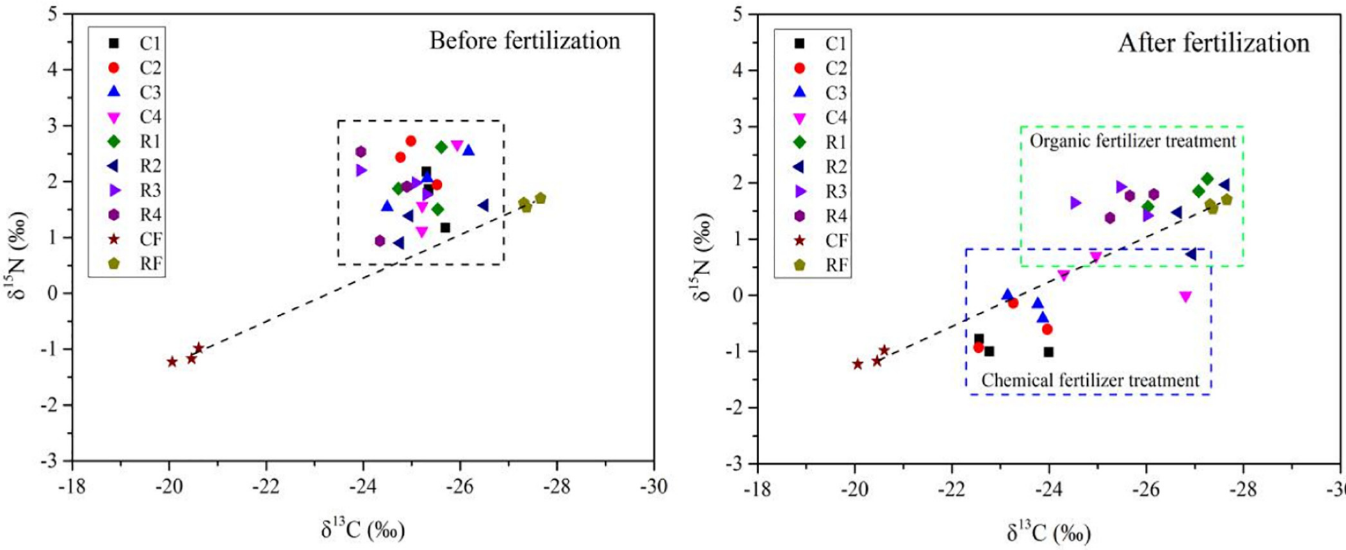

Fig. 5. Variation in $\delta^{13}C$ and $\delta^{15}N$ in the soils before and after fertilization. Treatment details refer to Table 1.

plantation soil exhibits acidic characteristics and a pH of approximately 4.0 to 5.5, which differs from the pH of other crops (Ruan et al., 2000; Yan et al., 2018). The soil pH in each treatment ranged from only 4.23 to 4.36, with an average of less than 4.50 before fertilization, and it gradually decreased after fertilization in the present study (Fig. 3). The results are consistent with many similar studies around the country; for example, the average soil pH of tea plantation was 4.68 nationally and ranged from 3.96 to 5.48 in different provinces of China (Yan et al., 2020). After the last fertilization, the soil pH in the C1 and C2 treatments had decreased to 3.88 and 3.95, respectively, the pH of C4 increased by 6.01% compared with that of C1, and the pH of R4 increased by 4.44% compared with that of R1. Compared with the pH in the compound fertilizer treatments, that in the rapeseed cake organic fertilizer treatments increased significantly by 2.19–4.29% under the corresponding fertilization rates. Therefore, both the fertilization rates and substitution of compound fertilizer with rapeseed cake organic fertilizer are appropriate methods for mitigating soil acidification in tea plantations.

Rapeseed cake organic fertilizer provided soils with more organic matter than compound fertilizers as observed in the TOM contents of the rapeseed cake organic fertilizer treatment groups, which were 8.04–21.14% higher than those in the corresponding compound fertilizer treatment groups. Richer soil TOM contents can promote soil microbial activity and the conversion of soil nutrients, which ultimately increase the tea plantation soil nutrient contents (Fan et al., 2016; Wang et al., 2016). The NH_4-N contents in the soil were significantly higher than the NO_3-N contents and the NH_4-N contents in the rapeseed cake organic fertilizer treatment groups were significantly higher than those in the compound fertilizer treatment groups (Fig. 3). Therefore, compared with compound fertilizer, rapeseed cake organic fertilizer is more conducive to the growth of tea trees, which prefer NH_4-N. Concomitantly, the NO_3-N contents in runoff water were higher than the NH_4-N contents (Fig. 3). NH_4-N in soil is usually adsorbed by soil particles and soil organic matter, and under aerobic soil conditions, NH_4-N can change rapidly to NO_3-N (Koper et al., 2010; Lteif et al., 2010).

. *Effects of fertilization types and rates on runoff nutrient loss*

In the study area, heavy rains over a short period of time usually lead to the rapid incorporation of NO_3-N from fertilizers into runoff water. Rapid nutrient losses are an important contributing factor for agricultural nonpoint pollution, especially in the study area, where heavy rainfall is concentrated in summer. Previous studies also showed that the amount of NH_4-N and NO_3-N loss is often highest throughout the year due to heavy rain after fertilization in agricultural watersheds (Udawatta and Motavalli, 2006; Y. Liu et al., 2012; Z.A. Liu et al., 2012). In this study, the correlations between rainfall and NH_4-N and NO_3-N contents also indicated that the average rainfall was the main cause of soil NH_4-N loss (Table 2); however, heavy rain over a short time represented an important cause of NO_3-N losses through runoff in the experimental field.

To further discuss the mass balance of water and nitrogen in the tea plantation, the losses of NH_4-N and NO_3-N during the entire fertilization experiment were calculated by the following formulas (Y. Liu et al., 2012; Z.A. Liu et al., 2012; Shan et al., 2015):

$$P = \sum_{i=1}^{n} C_i * Q_i \tag{1}$$

$$Q_i = K * R * A * 10^{-3} \tag{2}$$

where P represents the loss of NH_4-N and NO_3-N (kg/ha); C_i represents the content of NH_4-N and NO_3-N in runoff water in each rainfall event (mg/L); Q_i represents the amount of runoff from each rainfall event (L/m^2); R represents the amount of rainfall for each rainfall event

Table 2
Pearson correlation coefficients for runoff water indicators and rainfall.

Indicator	Treatment	Rainfall		
		Average	Cumulative	Maximum
NH_4-N	C1	0.618	−0.107	0.391
	C2	0.583	−0.202	0.295
	C3	0.641	−0.159	0.314
	C4	0.746	0.092	0.561
	R1	0.603	−0.192	0.289
	R2	0.565	−0.127	0.366
	R3	0.465	−0.109	0.166
	R4	−0.212	−0.421	−0.309
NO_3-N	C1	0.027	0.821	0.885*
	C2	0.023	0.832	0.898*
	C3	−0.025	0.791	0.895*
	C4	−0.237	0.650	0.817
	R1	−0.029	0.821	0.834
	R2	0.034	0.877	0.873
	R3	0.043	0.881*	0.900*
	R4	0.001	0.849	0.908*

Note: treatment details refer to Table 1. ** and * indicate significance at the 0.01 and 0.05 levels (two-tailed), respectively.

(mm); K represents the land surface runoff coefficient value (for tea plantations), which have a yellow brown soil vegetation surface, the runoff coefficient can be taken as 0.7 (Song et al., 2015); and A represents the catchment surface area of tea plantation (m^2) (Table 4).

N runoff losses were generally higher in treatments of compound fertilizer compared to rapeseed cake organic fertilizer throughout the year (Table 3). The calculation results showed that the cumulative loss of NH_4-N reached 3.79 kg/ha to 8.46 kg/ha from C4 to C1 and 3.40 kg/ha to 6.33 kg/ha from R4 to R1; the cumulative loss of NO_3-N reached 7.76 kg/ha to 24.95 kg/ha from C4 to C1 and 7.06 kg/ha to 18.93 kg/ha from R4 to R1 during the entire experiment. With decreasing fertilizer application rate, the losses of NH_4-N and NO_3-N in runoff water decreased correspondingly. The application of rapeseed cake organic fertilizer reduced NH_4-N runoff loss by 10.36–25.12% and NO_3-N runoff loss by 8.94–24.10% compared with compound fertilizer application.

Rainfall and fertilizer application are the most important factors determining N runoff losses. Soil erosion after fertilization caused by rainfall is the primary mechanism of N runoff loss in plantations (Franklin et al., 2007; Tan et al., 2013). Organic fertilizer application has been shown to increase organic matter content and increase water retaining capacity (Bulluck et al., 2002). Moreover, the nitrification capability of organic fertilizer is lower than that of inorganic fertilizer (Koper et al., 2010). Therefore, the nutrients in organic fertilizer may be released more slowly than traditional chemical fertilizer. A previous similar study also showed that the application of organic fertilizer reduced N runoff loss by 15.70–18.14% compared to conventional chemical fertilizer application (Shan et al., 2015). The results in this study are consistent with those of Shan et al. (2015) and suggest that rapeseed cake organic fertilizer application could increase the probability of N runoff losses compare with compound fertilizer application.

. Effects of fertilization types and rates on C and N sources and their isotopic compositions in soils

The application of fertilizer directly changed the C and N sources and their isotopic compositions in soils. Previous studies have also shown that the form of N application; i.e., organic versus inorganic or in combination, may affect the amount of C and N that becomes sequestered in soil to some extent (Moran et al., 2005; Ismaili et al., 2015). Although compound fertilizers and rapeseed cake organic fertilizers have the same nutrient contents under the same fertilization rates, they have different $\delta^{13}C$ and $\delta^{15}N$ values due to their unique properties. In this study, the average reference values of $\delta^{13}C$ and $\delta^{15}N$ for compound fertilizers were −20.38‰ and −1.13‰ while those of rapeseed cake organic fertilizers were −27.45‰ and 1.62‰, respectively. Obviously, there was a huge difference in the C and N isotopic composition of these two type fertilizers. The $\delta^{13}C$ and $\delta^{15}N$ of compound fertilizer, an artificial fertilizer commonly used in modern agriculture, are mainly determined by the mineral materials needed for fertilizer production. The $\delta^{13}C$ and $\delta^{15}N$ of rapeseed cake organic fertilizer, an organic fertilizer widely applied in the research area, are mainly determined by rape grown locally in Zhejiang Province, which is a typical source of plant-derived C and N. The more negative the $\delta^{13}C$ values and more positive $\delta^{15}N$ values in the rapeseed cake organic fertilization plots compared with the compound fertilization plots (Fig. 5) indicate that the isotopic fractionation of C and N in the soils might be dependent on the fertilizer type.

Table 3
NH_4-N and NO_3-N losses from February to October in the different treatments.

Treatment		C1	C2	C3	C4	R1	R2	R3	R4
NH_4-N loss (kg/ha)	Feb	0.19	0.21	0.20	0.19	0.19	0.21	0.21	0.18
	Apr	3.64	2.97	2.21	1.17	2.86	1.86	1.26	0.91
	Jun	1.87	1.40	1.33	1.27	1.37	1.50	1.42	1.13
	Aug	2.25	1.65	1.12	0.84	1.44	1.34	0.81	0.76
	Oct	0.51	0.47	0.41	0.32	0.47	0.43	0.44	0.42
NO_3-N loss (kg/ha)	Feb	0.29	0.34	0.32	0.30	0.33	0.30	0.33	0.31
	Apr	6.73	5.79	3.53	2.23	5.48	4.17	2.97	1.78
	Jun	7.16	5.93	3.80	2.42	4.68	3.92	3.04	2.31
	Aug	9.18	7.40	3.93	2.11	7.07	6.06	3.64	2.11
	Oct	1.59	1.26	0.88	0.68	1.38	0.92	0.67	0.56

Note: treatment details refer to Table 1.

Table 4
C and N contents before and after fertilization in the different treatments.

Treatment name	Before fertilization (g/kg)		After fertilization (g/kg)	
	C	N	C	N
C1	16.48	0.99	19.69	2.21
C2	17.15	1.28	20.72	2.26
C3	15.96	0.75	18.77	2.19
C4	18.25	0.86	19.10	2.04
R1	15.83	0.81	24.82	2.18
R2	16.70	1.24	23.33	2.31
R3	16.98	1.02	22.89	2.13
R4	16.35	1.01	21.04	2.07

Note: treatment details refer to Table 1.

To further determine the contribution of C or N in the soil from sources of fertilizer and soil, the pattern of conservation of stable isotope mass was used for calculation (Bird et al., 2008; Wang et al., 2018). The specific formulas were as follows:

$$C_{ferti}\ (\%) = \left(\delta^{13}C_{after} - \delta^{13}C_{before}\right) / \left(\delta^{13}C_{fertilizer} - \delta^{13}C_{before}\right) \times 100 \quad (3)$$

$$N_{ferti}\ (\%) = \left(\delta^{15}N_{after} - \delta^{15}N_{before}\right) / \left(\delta^{15}N_{fertilizer} - \delta^{15}N_{before}\right) \times 100 \quad (4)$$

$$C_{soil}\ (\%) = 100 - C_{ferti} \quad (5)$$

$$N_{soil}\ (\%) = 100 - N_{ferti} \quad (6)$$

where C_{ferti} and N_{ferti} represent the ratio of C and N sources from fertilizer, respectively; C_{soil} and N_{soil} represent the ratio of C and N sources from soil, respectively; $\delta^{13}C_{before}$ and $\delta^{15}N_{before}$ represent the C and N isotopic composition before fertilization, respectively; $\delta^{13}C_{after}$ and $\delta^{15}N_{after}$ represent the C and N isotopic composition after fertilization, respectively; and $\delta^{13}C_{fertilizer}$ and $\delta^{15}N_{fertilizer}$ represent the isotopic composition of compound fertilizer and rapeseed cake organic fertilizer, respectively.

The calculation results showed that for the compound fertilizer treatment groups, the C source from the compound fertilizer input in the soil accounts for 1.88–46.18%, with an average of 30.50% after fertilization, and for the rapeseed cake organic fertilizer, the C source accounts for 21.07–81.80%, with an average of 53.70% (Fig. 6). As the amount of fertilizer used decreased, the proportion of C input from fertilizers also gradually decreased and the C source input rates of the rapeseed cake organic fertilizer treatment groups were significantly higher than that of the compound fertilizer treatment groups. Moreover, the N source input from the compound fertilizer accounted for 53.82–98.12%, with an average of 69.50% after fertilization, and that from the rapeseed cake organic fertilizer accounted for 18.20–78.93%, with an average of 46.30% (Fig. 6). Compared with the C source input, the input ratios of the N to the soil from compound fertilizer were significantly higher than that of rapeseed cake organic fertilizer (Fig. 6). The difference in the ratio of C and N sources in the soil after fertilization might be because the compound fertilizer is a synthetic and fast nitrogen fertilizer that is mainly produced to provide the N required by the plants and thus inputs less C. However, the rapeseed cake organic fertilizer is a biomass fertilizer that also has a large amount of organic matter, which inputs a large amount of C to the soil. As a slow-release fertilizer, the N input of the rapeseed cake organic fertilizer is relatively small and slow. Similar studies also have shown that organic N from organic

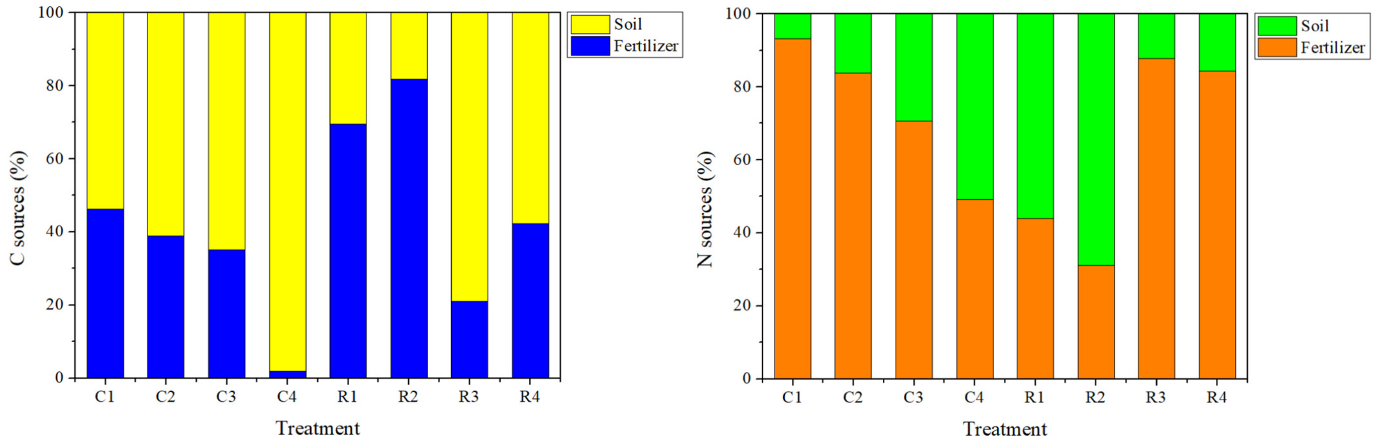

Fig. 6. Ratio of C and N sources from fertilizer and soil. Treatment details refer to Table 1.

fertilizers is more efficient than inorganic N from chemical fertilizers, especially in low-yield agricultural production systems (Busari et al., 2016).

Because tea is fertilized during multiple seasons throughout the year, the soil is able to quickly respond to the C and N changes caused by fertilizer inputs, which is a remarkable advantage that other crops lack. Due to the substantial differences in $\delta^{13}C$ and $\delta^{15}N$ compositions between compound fertilizer and rapeseed cake organic fertilizer, the soil exhibited significant isotope fractionation effects after one year of fertilization in this study, which allowed for the identification of fertilizer types applied in tea plantations based on the $\delta^{13}C$ and $\delta^{15}N$ isotope techniques. This phenomenon also reflects that the artificial compound fertilizers are mainly used as a source of N for growth while rapeseed cake organic fertilizers not only provide N but also help to preserve C in the soil. Thus, compound fertilizers and rapeseed cake organic fertilizers play different roles in influencing soil C and N cycling in tea plantations.

As an open tea plantation soil ecosystem, isotopic fractionation effects in N uptake by plants or isotopic fractionation effects during C and N transformation due to microbial reactions, including mineralization, nitrification, and denitrification will also occur (Oelbermann and Voroney, 2007; Ismaili et al., 2015). This phenomenon was also shown in the ratio of C and N sources in the soils from compound fertilizer or rapeseed cake organic fertilizer after fertilization (Fig. 6). Regarding the compound fertilizer treatments, with decreasing compound fertilizer amount, the ratio of C and N sources showed a very regular decline. However, for the rapeseed cake organic fertilizer treatments, this decrease was not as well defined. Rapeseed cake organic fertilizer can significantly increase the exogenous input of soil organic matter, which not only is more conducive to tea tree growth but also provides an excellent energy source for microbial activities. The C and N isotopic fractionation caused by plant uptake and microbial activity effects may be the main reason for the complex nonlinear relationship between the application amounts of rapeseed cake organic fertilizer and the ratio of C and N sources. Admittedly, other possible exogenous inputs to the soil include atmospheric sedimentation (Xue et al., 2013), tea garden irrigation (Choi et al., 2005) and soil microbial action (Tennakoon et al., 2019), which also play a role to some extent and need further exploration in the future.

. Conclusions

Rapeseed cake organic fertilization effectively buffered soil acidification, preserved the organic matter nutrient contents, promoted the growth of tea trees, and substantially reduced the N inputs to runoff compared with compound fertilization in the tea plantation. Heavy rainfall during the rainy season greatly contributed to the NH_4-N and NO_3-N inputs in runoff in the tea plantation, and management measures that substitute proportions of rapeseed cake organic fertilizer could be conducive to both maintaining soil fertility and reducing agricultural nonpoint pollution. Fertilization types and rates could be reflected in the $\delta^{13}C$ and $\delta^{15}N$ fractionations of tea plantation soils, and the $\delta^{13}C$ of the rapeseed cake organic fertilizer was significantly more negative than that of the compound fertilizer while the $\delta^{15}N$ was significantly more positive, suggesting a potential method of identifying the fertilizer type applied to tea plantations. Overall, considering the effects of C and N inputs to tea plantations, the application of rapeseed cake organic fertilizer should be a better strategy than the use of compound fertilizer.

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CRediT authorship contribution statement

Shaowen Xie: Investigation, Data curation, Writing - original draft. **Fen Yang:** Writing - review & editing. **Hanxiao Feng:** Investigation, Methodology. **Zhenzhen Yu:** Investigation, Project administration. **Chengshuai Liu:** Funding acquisition. **Chaoyang Wei:** Supervision. **Tao Liang:** Funding acquisition, Resources.

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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