



# Does dual reduction in chemical fertilizer and pesticides improve nutrient loss and tea yield and quality? A pilot study in a green tea garden in Shaoxing, Zhejiang Province, China

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## Abstract

Tea tree (*Camellia sinensis*) is a valuable and popular cash crop widely planted in tropical and subtropical areas of China. To increase tea yield and quality, high rates of chemical fertilizer and pesticide application have generally been used; however, increasing usage of fertilizers and pesticides does not always proportionally increase tea yield. Indeed, excessive nutrient inputs may cause serious agricultural non-point source pollution. A pilot study on dual reduction in fertilizers and pesticides was conducted in a green tea plantation in Shaoxing, Zhejiang Province, to explore the environmental effects of different fertilizer and pesticide managements (e.g., changes in soil properties and nutrient accumulation, nutrient inputs in runoff water) and to reveal the potential effects of the interaction of these two managements on tea yield and quality. Traditional formulas and rates of chemical fertilizers and pesticides were used as the baselines (100% usage); replacement with different proportions of organic fertilizer (i.e., 20%, 50% and 80%) and direct pesticide reductions of 30%, 50%, and 80% were tested. The results showed that proper management with organic fertilizer replacement can effectively mitigate soil acidification and nutrient deficiency in tea plantations, increase soil organic matter (OM) and ammonium nitrogen (NH<sub>4</sub>-N) contents, and promote tea yield and quality. Moreover, managements with organic fertilizer replacement can markedly reduce the inputs of ammonium nitrogen (NH<sub>4</sub>-N), nitrate nitrogen (NO<sub>3</sub>-N), total phosphorus (TP), and total potassium (TK) in runoff water. Soil nutrient accumulation was the highest while the runoff nutrient input was the lowest at 20% organic fertilizer replacement. Experimental spraying of bifenthrin and chlorfenapyr revealed that these pesticides were mainly trapped by the tea leaves and rarely entered the soil or water bodies. Although pesticide reduction treatments can effectively decrease pesticide residues in tea leaves, differences in pesticide residue between various treatments were not obvious due to the rapid degradation of pesticides. Multivariate analysis of variance showed that 50% of the variation in tea yield, bud density, polyphenols, and caffeine can be explained by interactions between fertilizers and pesticides. Combinations of 20% or 50% organic fertilizer replacement and 30% or 50% pesticide application reduction are appropriate for both mitigating nutrient loss and balancing tea yield and quality, especially the combination of 50% organic fertilizer replacement and 50% pesticide reduction, which produced the best results. This study demonstrates the feasibility of dual reductions in fertilizers and pesticides for mitigating environmental hazards while maintaining the yield and quality of tea.

**Keywords** Tea · Organic fertilizer · Pesticide residues · Environmental effects

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## Introduction

As a valuable and popular cash crop, tea (*Camellia sinensis*) is widely planted in the tropical and subtropical areas of China. In 2016, the total tea plantation area of China reached approximately 2.90 million ha, and the yield exceeded 2.4 million tons (NBSC 2016). The yield and quality of tea will directly affect the income of tens of thousands of tea farmers, and high fertilizer and pesticide application rates have generally been used to increase these parameters. Studies based on

surveys of tea farmers have shown that the average N fertilizer application rate is approximately 521 kg/ha, which far surpasses the required amount in the green tea plantations of Zhejiang Province, China (Ma et al. 2013). More seriously, recent research has shown that the N fertilizer application rate can even reach 1200 kg/ha in Chinese tea plantations (Wu et al. 2016). Regardless, the application of excess fertilizer and pesticides does not always proportionally increase yield. First, excessive fertilization can lead to severe soil acidification, and continually acidifying tea plantation soil will further lead to the accumulation of aluminum (Al) and accelerate the loss of phosphorus (P), potassium (K), and magnesium (Mg) nutrients from soils, which will then weaken the effect of fertilization (Wang et al. 1997; Ruan et al. 2013; Li et al. 2016; Yang et al. 2018). Second, considerable amounts of residual nitrogen and phosphorus from fertilizer and pesticides leach into runoff water and ultimately enter aquatic ecosystems, substantially contributing to the emission of pollutants into water bodies and soils (Sun et al. 2012; Maghanga and Kituyi 2013; Guo et al. 2014). Therefore, it is urgent to implement effective managements to reduce excessive fertilizer and pesticide inputs.

In Japan, high rates of nitrogen (N) fertilizer application are applied to tea fields, with nitrogen in the form of mineral and organic fertilizers being applied at rates as high as 450–1000 kg N/ha/year (Tokuda and Hayatsu 2000; Mishima et al. 2012). However, the N absorbed by mature tea plants is equivalent to only 200–300 kg N/ha<sup>-1</sup>/year, and the residual fertilizer leads to diffuse pollution and impedes the growth of tea plants (Nakasone and Yamamoto 2004; Oh et al. 2006). Similarly, in Kenya, Maghanga and Kituyi (2013) found that fertilizer application in tea gardens contributed to high nitrate levels in the receiving river. Many studies have documented fertilizer use in tea gardens and investigated alternative measures for reducing the amount of fertilizer applied. A study on vegetable farms in Virginia and Maryland (USA) used field experiments to examine the effects of organic and synthetic soil fertility amendments on soil physical and chemical properties (Bulluck et al. 2002), and the researchers found that compost application improved soil quality by increasing soil organic matter, total carbon, and cation exchange capacity. Mulla et al. (2007) reported that in Minnesota (USA), alternative agricultural practices, such as organic production, have been promoted as eco-friendly due to the reduced associated risks of water pollution. An investigation in Italy showed that fertilizer management with organic fertilizers can be a valuable alternative to traditional mineral fertilization of nectarine trees cultivated in the Po Valley (Baldi et al. 2016). Therefore, it can be hypothesized that alternative methods involving organic fertilizer can reduce loss of nutrients such as nitrogen and phosphorus while increasing soil fertility in the tea gardens of Southern China.

The prevention and control of insect pests is also indispensable for guaranteeing the yield and quality of leaf-harvested

commercial crops in tea plantations. The false-eye leafhopper *Empoasca vitis* is the predominant insect pest and is widely distributed in tea plantations of Southern China (Feng et al. 2004; Pu et al. 2005), particularly in the green tea plantations of Zhejiang. These pests move among plants to taste and suck from fresh, tender leaves, resulting in dramatic decreases in yield and quality due to curling and parching of the damaged leaves (Miao and Han 2007). Although pesticide spraying can effectively kill these insect pests and mitigate tea yield and quality loss, excessive spraying of pesticides may lead to pesticide residues in the tea and ultimately affect tea sales and harm human health. Ensuring that such residue is as harmless as possible to the tea garden environment without exceeding the limit is a key issue. At present, the amount of pesticide applied by tea farmers is generally based on guidelines from the pesticide's manufacturer. However, these guidelines are usually set at a very broad range; thus, specific application rates should be based on the specific conditions of the tea production area. To date, there is relatively little research on the environmental impact of pesticide application on pesticide residues in tea leaves, soils, and water bodies in tea gardens, and rational pesticide management that can balance pesticide residues in tea products with tea yield and quality is required.

Fertilizers and pesticides are two important production materials in the process of modern tea planting, but current research on the effects of fertilizer and pesticide reduction managements on soil properties, runoff water, tea yield, and quality is rather limited. The detrimental impact of tea cultivation on the environment can be effectively reduced by appropriate managements to decrease fertilizer and pesticide application, and understanding the possible interactions between different fertilizer and pesticide managements using rational planning of fertilizer and pesticide applications is crucial. Research regarding the management of fertilizer and pesticide reduction is not only helpful for lowering the cost of tea cultivation but also beneficial for decreasing the potential risk of environmental pollution in tea plantations. The objectives of this study were as follows: (1) to explore the effects of various ratios of organic fertilizer replacement on soil properties, nutrient inputs to runoff water, and tea yield and quality; (2) to illustrate the environmental effects of different ratios of pesticide reduction on the pesticide residues in tea leaves, soils, and runoff water; and (3) to reveal the potential effects of interactions between dual reductions in fertilizer and pesticides on tea yield and quality.

## Materials and methods

### Experimental site description

The experimental tea field is located at Yucha Village near the suburb of Shaoxing City, Zhejiang Province, China (Fig. 1). The area is dominated by a hilly landscape with an average

elevation of 166 m, and the soil is mostly yellow brown soil. The annual average temperature is 15 °C, and the annual average precipitation is between 1200 and 1400 mm, which categorizes the area as having a subtropical humid monsoon climate. Spring is warm and humid, and summer is hot. Heavy rainfall occurs in a short time during summer. The tea trees used for the experiment in the plantation were more than 10 years old, and this is one of the largest-scale green tea planting fields in Zhejiang Province. The pilot experimental tea plots were selected within the area of the field mentioned above, in which tea trees have been naturally grown for more than 5 years without any manual fertilizer or pesticide treatments, guaranteeing the reliability of the study in terms of eliminating the influence of external factors.

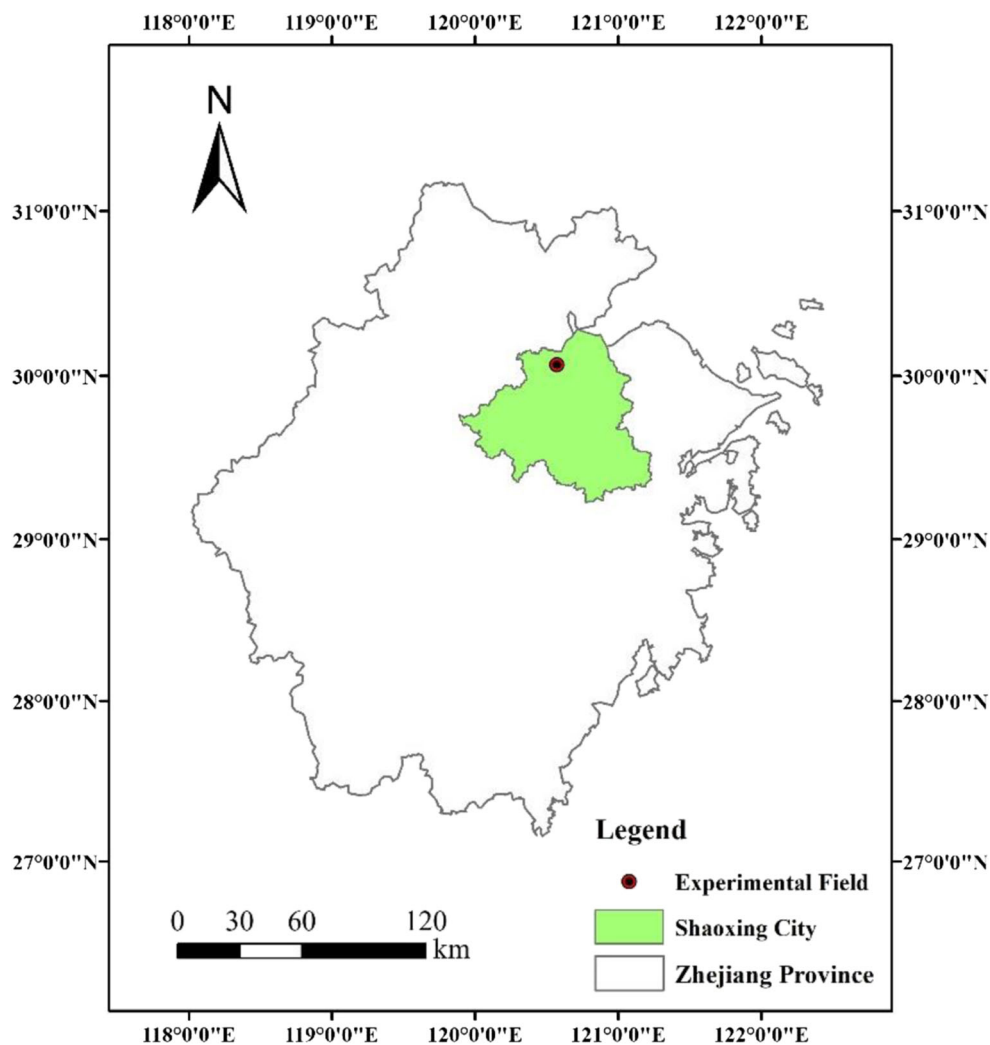
### Experimental design and statistical analysis

The experiment was performed with two factors, namely chemical fertilizers and pesticides, each with four levels, in a non-complete group design. The baseline usage doses of

fertilizer and pesticides were used as the 100% control group; different ratios of organic fertilizer replacements or reductions in pesticide usage were employed as the treatment groups. A total of ten treatments were included, and each treatment was repeated three times (Table 1). All the experimental plots, with an area of 21 m<sup>2</sup> (3 m × 7 m), were randomly arranged on an 18° slope with a length parallel to the slope. A 1-m buffer and an isolation barrier were placed between adjacent plots to prevent water intrusion. In addition, runoff buckets were set according to the flow direction of runoff water at the bottom of each experimental plot to collect runoff water.

Reductions in fertilizer were carried out by using organic fertilizer replacement, and the fertilization treatments included four different levels: the baseline, 100% chemical fertilizer (F<sub>b</sub>); 20% organic fertilizer replacement (F<sub>1</sub>); 50% organic fertilizer replacement (F<sub>2</sub>); and 80% organic fertilizer replacement (F<sub>3</sub>). The baseline of 100% chemical fertilization was designed according to the annual pure nitrogen application rate of 450 kg/hm<sup>2</sup>, which was obtained through questionnaire surveys of local tea farmers and tea production companies. In

**Fig. 1** The location of the experimental site



**Table 1** Design of the *dual-reduction* chemical fertilizer and pesticide treatments

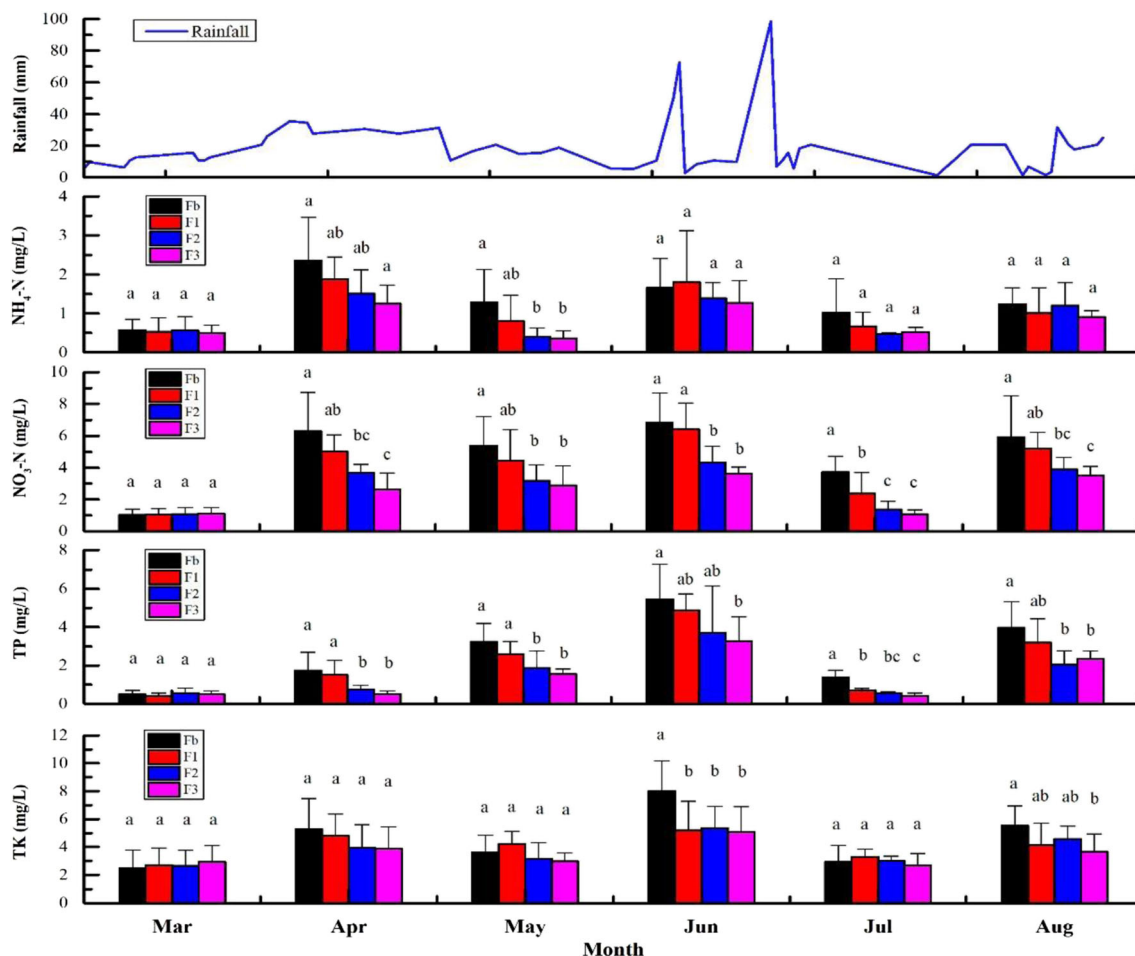
| Treatment name | Fertilizer treatment                    | Pesticide treatment |
|----------------|---|---------------------|
| Treatment 1    | 100% chemical fertilizer (Fb)           | 100% pesticide (Pb) |
| Treatment 2    | 20% organic fertilizer replacement (F1) | 70% pesticide (P1)  |
| Treatment 3    |   | 50% pesticide (P2)  |
| Treatment 4    |   | 20% pesticide (P3)  |
| Treatment 5    | 50% organic fertilizer replacement (F2) | 70% pesticide (P1)  |
| Treatment 6    |   | 50% pesticide (P2)  |
| Treatment 7    |   | 20% pesticide (P3)  |
| Treatment 8    | 80% organic fertilizer replacement (F3) | 70% pesticide (P1)  |
| Treatment 9    |   | 50% pesticide (P2)  |
| Treatment 10   |   | 20% pesticide (P3)  |

contrast to the traditional method of replacing nitrogen fertilizer and considering the comprehensive nutrient element demands of tea growth, a method of completely replacing N, P<sub>2</sub>O<sub>5</sub>, and K<sub>2</sub>O nutrients was adopted in this study. The effects of replacement application of organic fertilizers on soil nutrient inputs and runoff-water nutrient inputs under the conditions of equal nutrient input were then investigated. N:P<sub>2</sub>O<sub>5</sub>:K<sub>2</sub>O was applied in the tea plantation at a ratio of 3:1:2 (N = 450 kg/hm<sup>2</sup>, P<sub>2</sub>O<sub>5</sub> = 150 kg/hm<sup>2</sup>, K<sub>2</sub>O = 300 kg/hm<sup>2</sup>), and the types of fertilizer used included urea, calcium superphosphate, potassium sulfate, and compound fertilizer, which the tea farmers had already generally been using. The approximate specific nutrient contents in the fertilizers were as follows: urea (N = 46%), calcium superphosphate (P<sub>2</sub>O<sub>5</sub> = 12%), potassium sulfate (K<sub>2</sub>O = 60%), and compound fertilizer (N = 150 g/kg, P<sub>2</sub>O<sub>5</sub> = 150 g/kg, K<sub>2</sub>O = 150 g/kg). The type of organic fertilizer used in this study was rape seed cake, which is partly used as an organic fertilizer in Zhejiang Province. The NPK contents in the rape-seed cake were N = 52.7 g/kg, P<sub>2</sub>O<sub>5</sub> = 6.9 g/kg, and K<sub>2</sub>O = 6.1 g/kg, respectively. All the fertilizers were applied in the band furrows at a depth of 15 cm between two rows of tea plants and then covered with topsoil. Fertilization was performed at the end of March, May, and July; thus, the proportion of the year with fertilization was 5:3:2. The effects of fertilization treatments on soil properties were obtained by analyzing monthly changes in soil properties before and after fertilization. Ammonium nitrogen (NH<sub>4</sub>-N), nitrate nitrogen (NO<sub>3</sub>-N), total phosphorus (TP), and total potassium (TK) contents in tea plantation runoff water were measured monthly. Additionally, daily rainfall was recorded throughout the fertilization process (Fig. 2). Correlations between runoff contents and average rainfall on rainy days during the month, monthly cumulative rainfall, and monthly maximum rainfall were analyzed to explore the relationships between these indices.

Field surveys were conducted before the spraying of any pesticides, which showed that the average number of false-eye leafhoppers in the tea garden was 42 individuals per hundred leaves in June and 44 per hundred

leaves in August. Thus, the risk of pests in experimental tea garden persisted. Four levels of pesticide management were also established: 100% pesticide (Pb), 70% pesticide (P1), 50% pesticide (P2), and 20% pesticide (P3). The baseline treatment of 100% pesticide was obtained from local tea farmers and tea production companies by detailed surveys, and the pesticides used were bifenthrin and chlorfenapyr, which are specifically employed to prevent false-eye leafhopper and are generally used in the green tea plantations of Zhejiang Province. Bifenthrin was sprayed at the beginning of June, and chlorfenapyr was sprayed at the beginning of August, considering summer is the peak season of false-eye leafhopper abundance. The formulations used were a 10% bifenthrin emulsifiable concentrate and a 240 g/L chlorfenapyr suspension concentrate, which are widely utilized by tea farmers. A backpack manual pressure atomizer was employed to spray the pesticide. To further study the environmental impact of pesticides, the pesticide residues in tea leaves, soil, and runoff water were measured. All the specific types and dosages of fertilizers and pesticides are presented in Tables 2 and 3, respectively, and to reduce the influence of other factors, all other managements used to ensure the normal growth of the tea trees were consistent. The field experiment started in February 2017 and continued for an entire year, covering a full year of fertilizer and pesticide application and tea harvesting cycles.

Samples of the 0–20-cm soil layer were collected from 10 cm outside the drip line (fertilization point) in the treatments. The collected soil samples were dried, and soil indicators were determined after mixing and sifting. The collected runoff water was filtered, stored in a refrigerator, and transported to the laboratory for testing as soon as possible. The pH of soils was determined in 1:5 soil:water suspension solutions using a pH meter, soil organic matter (OM) was measured using an elemental analyzer, and soil NH<sub>4</sub>-N and NO<sub>3</sub>-N were evaluated with an automatic chemical analyzer.



**Fig. 2** The rainfall and NH<sub>4</sub>-N, NO<sub>3</sub>-N, TP and TK contents from runoff water. Fb is the 100% chemical fertilizer treatment, F1 is the 20% organic fertilizer replacement treatment, F2 is the 50% organic fertilizer

replacement treatment, and F3 is the 80% organic fertilizer replacement treatment. Values followed by different letters are significantly different ( $P \leq 0.05$ )

Available phosphorus (Olsen P) and available potassium (Olsen K) were extracted with 2% (NH<sub>4</sub>)<sub>2</sub>CO<sub>3</sub> and then determined by inductive coupled plasma optical emission spectrometry (ICP-OES). This method allowed simultaneous measurement of Olsen P and Olsen K and yields reliable results (Tian 1997). Runoff water TP and TK were also determined by ICP-OES after digestion with acid (nitric acid:hydrofluoric acid:perchloric acid = 3:3:1). All pesticide residues in tea

leaves, soil, and runoff water were measured by gas chromatography, with a detection limit in leaves of 0.004 mg/kg and soil of 0.001 mg/kg. All tea samples were measured for bud density (tea bud number within 0.1 m<sup>2</sup>) during germination and fresh weight yield during harvest in each plot. In addition, tea yield, bud density, polyphenols, caffeine, water extracts, and amino acids, which are six useful indices for evaluating the yield and quality of tea, were determined.

**Table 2** Types and application amounts of fertilizers

| Fertilizers                        | Type and application amount of fertilizer (kg/hm <sup>2</sup> ) |      |                        |                   |                          |
|------------------------------------|---|------|------------------------|-------------------|--------------------------|
|                                    | Compound fertilizer   | Urea | Calcium superphosphate | Potassium sulfate | Rape seed cake (organic) |
| 100% chemical fertilizer           | 600   | 782  | 500                    | 350               | –                        |
| 20% organic fertilizer replacement | –   | 761  | 1049                   | 480               | 1904                     |
| 50% organic fertilizer replacement | –   | 478  | 995                    | 450               | 3928                     |
| 80% organic fertilizer replacement | –   | 191  | 870                    | 421               | 6207                     |



**Table 3** Types and application amounts of pesticides

| Pesticides     | Type and application amount of pesticides (mL/hm <sup>2</sup> ) |              |
|----------------|---|--------------|
|                | Bifenthrin  | Chlorfenapyr |
| 100% pesticide | 150   | 750          |
| 70% pesticide  | 105   | 525          |
| 50% pesticide  | 75  | 375          |
| 20% pesticide  | 30  | 150          |

All statistical analyses were carried out using SPSS V20 for Windows. One-way ANOVA with the Student–Newman–Keuls test was performed to detect significant differences in various factors between treatments and to reveal the single and interactive effects of fertilizer replacement and pesticide reduction on the yield and quality of tea ( $P \leq 0.05$ ). Correlations were assessed by the Pearson correlation, and multivariate ANOVA was used for interaction analysis ( $P \leq 0.05$ ).

## Results

### Effects of different fertilizer treatments on soil properties

All key physical and chemical characteristics of the soil of the tea plantation were measured at the beginning of the experiment and before fertilization (i.e., in March); the measurements are presented in Table 4: pH ( $4.29 \pm 0.01$ ), OM ( $18.17 \pm 0.45$  g/kg), NH<sub>4</sub>-N ( $6.48 \pm 0.31$  mg/kg), NO<sub>3</sub>-N ( $5.79 \pm 0.43$  mg/kg), Olsen P ( $84.28 \pm 4.02$  mg/kg), and Olsen K ( $94.93 \pm 4.57$  mg/kg). No significant differences in various soil properties were observed between treatments, which confirmed that the experiment had consistent initial conditions. After a full year of fertilization, substantial changes were observed in the soil of the tea plantation. At the end of August, after the last fertilization, the soil characteristics were as follows: pH ( $4.03 \pm 0.07$ ), OM ( $23.88 \pm 0.71$  g/kg), NH<sub>4</sub>-N ( $93.86 \pm 32.13$  mg/kg), NO<sub>3</sub>-N ( $34.32 \pm 13.36$  mg/kg), Olsen P ( $106.35 \pm 14.14$  mg/kg), and Olsen K ( $115.50 \pm 17.23$  mg/kg). Thus, soil nutrients after fertilization were significantly higher than those before fertilization due to the addition of artificial fertilizer. Except for pH, soil factors were significantly improved by fertilization: OM content, Olsen P, and Olsen K increased by 31.4%, 26.2%, and 21.7%, respectively. The increases in NH<sub>4</sub>-N and NO<sub>3</sub>-N were the largest, being 14 and 6 times greater than those before fertilization, respectively. Compared with the Fb treatment, the treatments with organic fertilizer replacement, namely F1, F2, and F3, improved the soil pH, OM, NH<sub>4</sub>-N, NO<sub>3</sub>-N, Olsen P, and Olsen K to different degrees. After each fertilization, the pH

value in the F1, F2, and F3 treatments was 0.8–1.8%, 1.7–4.8%, and 2.2–6.0% higher than that in the Fb treatment, and the OM content in the F1, F2, and F3 treatments was 1.5–13.8%, 7.0–54.7%, and 4.3–39.1% higher than that in the Fb treatment, respectively. There were significant differences among the F1, F2, F3, and Fb treatments, and the most significant increase occurred in the F3 treatment.

Regarding soil NH<sub>4</sub>-N and NO<sub>3</sub>-N, the NH<sub>4</sub>-N content in the F1, F2, and F3 treatments was 8.4–79.6%, 1.0–148.3%, and 0.9–113.9% higher than that in the Fb treatment, and the NO<sub>3</sub>-N content in the F1, F2, and F3 treatments was 16.2–53.8%, 35.0–153.4%, and 20.8–51.8% higher than that in the Fb treatment, respectively. Significant differences between the F1, F2, F3, and Fb treatments were observed, except for between F1 and F3. Unlike pH and OM, the largest increase in NH<sub>4</sub>-N and NO<sub>3</sub>-N was found in the F2 treatment. When the ratio of replacement with organic fertilizer exceeded 50% and 80%, the NH<sub>4</sub>-N and NO<sub>3</sub>-N in the soil did not increase; instead, they fell to levels comparable to those in the F1 treatment. Similar trends also occurred in terms of Olsen K; specifically, the Olsen K content in the F1, F2, and F3 treatments was 3.3–30.7%, 15.3–44.8%, and 10.3–29.8% higher than that in the Fb treatment, respectively. Significant differences were also observed between the F1, F2, F3, and Fb treatments, but not between the F1 and F3 treatments. The Olsen K content in the soil did not continue to increase with an increasing ratio of replacement with organic fertilizer, and the Olsen P content in the organic fertilizer replacement treatments was slightly higher than that in the Fb treatment.

### Effects of different fertilizer treatments on nutrient inputs in runoff water

Before fertilization, in March, the untreated initial concentrations of runoff water were approximately the following: NH<sub>4</sub>-N ( $0.54 \pm 0.03$  mg/L), NO<sub>3</sub>-N ( $1.06 \pm 0.03$  mg/L), TP ( $0.49 \pm 0.06$  mg/L), and TK ( $2.69 \pm 0.19$  mg/L). After fertilization, in August, the concentrations of NPK in runoff increased to approximately the following: NH<sub>4</sub>-N ( $1.22 \pm 0.24$  mg/L), NO<sub>3</sub>-N ( $4.64 \pm 1.13$  mg/L), TP ( $2.88 \pm 0.88$  mg/L), and TK ( $4.47 \pm 0.80$  mg/L). Fertilizer application significantly increased NPK nutrient inputs in runoff water after each fertilization. Moreover, the NPK content in the organic fertilizer replacement treatments (F1, F2, and F3) was significantly lower than that in the 100% chemical fertilizer treatment (Fb). In general, the nitrate content in tea plantation runoff was dominated by NO<sub>3</sub>-N, and the nitrate nitrogen content was significantly higher than the NH<sub>4</sub>-N content after fertilization. Specifically, the NH<sub>4</sub>-N in runoff water in the organic fertilizer replacement treatments was lower than that in the Fb treatment and was most pronounced in May. The order of average concentrations of runoff NH<sub>4</sub>-N in the treatments was Fb > F1 > F2 > F3; specifically, that in the F1, F2, and F3 treatments was

**Table 4** Effects of fertilization treatments on soil indicators

| Indicator                  | Treatment | March          | April           | May             | June             | July            | August          |
|----------------------------|-----------|----------------|-----------------|-----------------|------------------|-----------------|-----------------|
| pH                         | Fb        | 4.28 ± 0.14a   | 4.13 ± 0.06a    | 4.11 ± 0.08a    | 4.00 ± 0.09b     | 3.97 ± 0.07b    | 3.95 ± 0.07b    |
|                            | F1        | 4.29 ± 0.10a   | 4.19 ± 0.13a    | 4.19 ± 0.11a    | 4.07 ± 0.10b     | 4.04 ± 0.12b    | 3.98 ± 0.08b    |
|                            | F2        | 4.31 ± 0.14a   | 4.20 ± 0.16a    | 4.20 ± 0.15a    | 4.19 ± 0.19a     | 4.15 ± 0.09a    | 4.08 ± 0.11a    |
|                            | F3        | 4.29 ± 0.07a   | 4.22 ± 0.08a    | 4.23 ± 0.08a    | 4.24 ± 0.09a     | 4.19 ± 0.05a    | 4.10 ± 0.09a    |
| OM (g/kg)                  | Fb        | 17.99 ± 2.61a  | 19.50 ± 2.94c   | 15.29 ± 1.36c   | 18.66 ± 3.40b    | 22.41 ± 4.83a   | 23.14 ± 1.98b   |
|                            | F1        | 17.79 ± 3.48a  | 22.19 ± 2.62bc  | 19.47 ± 2.47b   | 20.69 ± 5.96b    | 26.06 ± 4.02a   | 23.48 ± 3.03ab  |
|                            | F2        | 18.82 ± 4.52a  | 23.77 ± 4.43b   | 21.96 ± 2.64ab  | 25.28 ± 4.07a    | 21.31 ± 2.50a   | 24.75 ± 3.26a   |
|                            | F3        | 18.08 ± 4.41a  | 27.12 ± 3.27a   | 22.94 ± 4.34a   | 28.87 ± 4.98a    | 23.54 ± 3.53a   | 24.13 ± 4.27a   |
| NH <sub>4</sub> -N (mg/kg) | Fb        | 6.14 ± 1.27a   | 10.49 ± 2.82a   | 16.21 ± 6.01b   | 45.90 ± 19.09b   | 25.97 ± 17.74b  | 50.61 ± 25.26b  |
|                            | F1        | 6.85 ± 0.98a   | 11.37 ± 3.91a   | 19.88 ± 3.39ab  | 77.03 ± 34.27ab  | 50.81 ± 25.91ab | 90.90 ± 46.97ab |
|                            | F2        | 6.58 ± 1.06a   | 10.60 ± 3.43a   | 23.89 ± 6.42a   | 88.15 ± 37.20a   | 76.22 ± 18.70a  | 125.64 ± 41.19a |
|                            | F3        | 6.33 ± 1.08a   | 10.58 ± 2.75a   | 21.33 ± 3.07ab  | 56.99 ± 14.94ab  | 66.95 ± 45.35a  | 108.28 ± 50.19a |
| NO <sub>3</sub> -N (mg/kg) | Fb        | 5.23 ± 0.64a   | 6.77 ± 2.04b    | 7.41 ± 2.38a    | 16.07 ± 4.15a    | 24.83 ± 20.71a  | 20.83 ± 5.92b   |
|                            | F1        | 6.02 ± 0.68a   | 8.88 ± 1.34a    | 7.60 ± 2.01a    | 18.67 ± 13.95a   | 27.25 ± 12.66a  | 32.03 ± 12.72b  |
|                            | F2        | 6.21 ± 0.71a   | 9.14 ± 1.17a    | 7.44 ± 2.00a    | 27.12 ± 11.06a   | 23.01 ± 12.01a  | 52.79 ± 19.97a  |
|                            | F3        | 5.70 ± 1.25a   | 8.18 ± 1.45ab   | 6.88 ± 1.15a    | 21.30 ± 15.08a   | 20.58 ± 8.93a   | 31.63 ± 16.28b  |
| Olsen P (mg/kg)            | Fb        | 79.95 ± 25.71a | 87.50 ± 14.49a  | 60.75 ± 15.77a  | 87.03 ± 29.40a   | 68.46 ± 19.97a  | 85.49 ± 29.38a  |
|                            | F1        | 82.66 ± 28.36a | 95.78 ± 19.40a  | 67.76 ± 14.59a  | 105.52 ± 22.23a  | 70.59 ± 25.08a  | 113.98 ± 32.38a |
|                            | F2        | 89.43 ± 18.91a | 100.07 ± 18.53a | 73.00 ± 25.62a  | 109.01 ± 36.15a  | 91.84 ± 20.62a  | 116.04 ± 34.02a |
|                            | F3        | 85.09 ± 15.55a | 96.90 ± 22.22a  | 86.31 ± 27.60a  | 104.40 ± 17.05a  | 89.73 ± 29.54a  | 109.89 ± 40.22a |
| Olsen K (mg/kg)            | Fb        | 99.06 ± 17.98a | 101.47 ± 25.44a | 74.67 ± 17.69b  | 87.01 ± 18.47b   | 76.46 ± 16.99c  | 91.45 ± 11.19b  |
|                            | F1        | 94.40 ± 18.42a | 104.83 ± 28.70a | 85.98 ± 10.23ab | 100.89 ± 28.33ab | 89.31 ± 19.16bc | 119.48 ± 23.68a |
|                            | F2        | 97.54 ± 15.05a | 117.02 ± 31.01a | 95.31 ± 15.71a  | 121.03 ± 33.48a  | 111.76 ± 17.42a | 132.42 ± 25.95a |
|                            | F3        | 88.72 ± 19.58a | 111.88 ± 27.68a | 91.12 ± 8.36a   | 103.44 ± 20.53ab | 97.88 ± 17.00ab | 118.67 ± 19.20a |

Fb is the 100% chemical fertilizer treatment, F1 is the 20% organic fertilizer replacement treatment, F2 is the 50% organic fertilizer replacement treatment, and F3 is the 80% organic fertilizer replacement treatment. Different letters following values indicate significant differences ( $P \leq 0.05$ ) between different treatments

37.4%, 69.1%, and 71.9% lower than that in the Fb treatment, respectively. The average NO<sub>3</sub>-N content in runoff water also followed the pattern Fb > F1 > F2 > F3. Organic fertilizer replacement resulted in a significantly lower NO<sub>3</sub>-N content than in the Fb treatment, and the most significant effect occurred in July. The NO<sub>3</sub>-N in the F1, F2, and F3 treatments was 36.4%, 63.1%, and 71.0% lower than that in the Fb treatment, respectively.

The trends of TP and TK were consistent with those of NH<sub>4</sub>-N and NO<sub>3</sub>-N. Specifically, after fertilization, the TP and TK in runoff water were significantly higher than those before fertilization, and they were significantly lower in the organic fertilizer replacement treatments than in the 100% chemical fertilizer treatment. The TP in the F1, F2, and F3 treatments was 10.7–49.8%, 32.1–60.2%, and 40.1–70.2% lower than that in the Fb treatment, respectively, after each fertilization in April, June, and August. Similarly, the TK in the F1, F2, and F3 treatments was 8.8–35.2%, 17.0–33.3%, and 26.5–36.8% lower than that in the Fb treatment after each fertilization in April, June, and August. Thus, using organic

fertilizer significantly reduced the phosphorus and potassium contents in tea plantation runoff.

During the experiments, the maximum runoff NH<sub>4</sub>-N, NO<sub>3</sub>-N, TP, and TK in all treatments usually appeared in June, which was closely related to rainfall in the study area (Fig. 2). Positive correlations were found between NH<sub>4</sub>-N, NO<sub>3</sub>-N, TP, and TK contents and monthly cumulative and maximum rainfall. Specifically, the correlation coefficients between NH<sub>4</sub>-N, NO<sub>3</sub>-N, and TK and monthly cumulative rainfall were greater than those between NH<sub>4</sub>-N, NO<sub>3</sub>-N, and TK and monthly maximum rainfall, while the correlation coefficient between TP and monthly maximum rainfall was greater than that between TP and monthly cumulative rainfall (Table 5).

### Effects of different pesticide treatments on tea leaves, soil, and runoff water pesticide residues

The strongest impact of pesticide management on the tea plantation environment manifests as pesticide residues on

**Table 5** Pearson correlation coefficients for runoff water indicators and rainfall

| Indicator          | Treatment | Rainfall |            |         |
|--------------------|-----------|----------|------------|---------|
|                    |           | Average  | Cumulative | Maximum |
| NH <sub>4</sub> -N | CF        | 0.767    | 0.620      | 0.281   |
|                    | OF1       | 0.404    | 0.885*     | 0.581   |
|                    | OF2       | 0.309    | 0.831      | 0.527   |
|                    | OF3       | 0.254    | 0.860      | 0.626   |
| NO <sub>3</sub> -N | CF        | 0.532    | 0.941*     | 0.704   |
|                    | OF1       | 0.403    | 0.938*     | 0.767   |
|                    | OF2       | 0.478    | 0.882*     | 0.640   |
|                    | OF3       | 0.344    | 0.778      | 0.599   |
| TP                 | CF        | -0.086   | 0.715      | 0.806   |
|                    | OF1       | 0.013    | 0.794      | 0.850   |
|                    | OF2       | -0.066   | 0.742      | 0.877   |
|                    | OF3       | -0.150   | 0.694      | 0.787   |
| TK                 | CF        | 0.106    | 0.968**    | 0.898*  |
|                    | OF1       | 0.580    | 0.960**    | 0.788   |
|                    | OF2       | -0.039   | 0.897*     | 0.820   |
|                    | OF3       | 0.174    | 0.985**    | 0.920*  |

\*Significant at the 0.05 level (two-tailed)

\*\*Significant at the 0.01 level (two-tailed)

leaves, which is also a great concern for tea producers and consumers. In general, pesticide residues in the three major carriers, namely tea, soil, and runoff water, showed the following rank order: tea leaf residues > soil residues > runoff water residues (Table 6). Specifically, the experimental results indicated that tea leaf residues were 0.08–0.71 mg/kg in all bifenthrin treatments and 0.02–0.35 mg/kg in all chlorfenapyr treatments between the 1st and 10th days after the pesticide was sprayed. The soil residues were 0.001–0.005 mg/kg in all bifenthrin treatments and 0.001–0.012 mg/kg in all chlorfenapyr treatments during the experiment with pesticide spraying. Although the residues of runoff water in all bifenthrin and chlorfenapyr treatments were only 0.001–0.002 mg/kg, most were not detectable. Clearly, pesticide residues were mainly trapped by tea leaves, and very little entered the soil and water bodies. However, further analysis actually revealed that all pesticide levels in tea leaves were far below the maximum pesticide residue in food limits prescribed by the national food safety standards of 5 mg/kg for bifenthrin and 20 mg/kg for chlorfenapyr (GB 2763-2016), meaning that all pesticide treatments are safe to use.

Pesticide reduction treatments can effectively reduce pesticide residues in tea leaves; however, the pesticide residue changes will decrease over time. For the bifenthrin treatments, the differences in the residual

amount of pesticide residue in tea leaves between four pesticide treatment levels on the same day were significantly decreased, and the pesticide residues on the leaf surface gradually decreased with the increase in the pesticide reduction gradient. In addition, the residual amounts in the 100% bifenthrin treatment (PB) were significantly higher than those in the other pesticide reduction treatments, and tea leaf residues in the PB2 and PB3 treatments were 38.0–42.1% and 64.8–67.9% lower than those in the PB treatment, respectively. For the chlorfenapyr treatments, there were significant differences in pesticide residues between the four different treatments but only on the 1st and 3rd days of application, as the differences were not significant on the 7th and 10th days of application. On the 1st day, the residues in the PC2 and PC3 treatments were 40.0% and 71.4% lower than those in the PC treatment, but those in the PC1 and PC treatments were not significantly different. On the 3rd day, the residues in the PC2 and PC3 treatments were 31.3% and 62.5% lower than those in the PC treatment, whereas there were no significant differences on the other days.

The degradation of pesticide residues is another important indicator that needs attention, and it will directly indicate the duration of pesticide use. Considering that there were very few pesticide residues in soil and runoff water, the degradation of pesticide residues was analyzed only on tea leaves. The degradation kinetics of the bifenthrin and chlorfenapyr residues on tea leaves were determined by plotting residue concentration against time, and the maximum squares of correlation coefficients were used to determine the equations of best-fit curves (Tewary et al. 2005). For all treatments, exponential relationships corresponding to a first-order rate equation were found, and the rate equation was calculated from the first-order rate equation:

$$C_t = C_0 \cdot e^{-k}$$

where  $C_t$  is the concentration of the pesticide residue at time  $t$ ,  $C_0$  is the initial concentration, and  $k$  is the rate constant in per day. The half-lives ( $t_{1/2}$ ) were determined from the  $k$  value for each treatment as  $t_{1/2} = \ln 2/k$ .

All the dynamic equations, correlation coefficients and half-lives are presented in Table 7. The correlation coefficients of bifenthrin treatments were  $R^2 = 0.896–0.983$ , and the half-lives of pesticide dissolution were 4.87–6.13 days. The correlation coefficients of chlorfenapyr treatments were  $R^2 = 0.782–0.928$ , and the half-lives of pesticide dissolution were 2.56–5.55 days. The half-lives of chlorfenapyr were slightly lower than those of bifenthrin, indicating that chlorfenapyr degraded more rapidly than bifenthrin. The short half-life of chlorfenapyr also indicated that the pesticide can be completely degraded before the tea is harvested, thereby minimizing its environmental impact.



**Table 6** Residues of tea leaves, soil, and runoff water after spraying pesticides

|                                     | 1st day         | 3rd day        | 7th day        | 10th day       |
|-------------------------------------|-----------------|----------------|----------------|----------------|
| Tea leaf pesticide residues (mg/kg) |                 |                |                |                |
| PB                                  | 0.71 ± 0.09a    | 0.53 ± 0.04a   | 0.38 ± 0.10a   | 0.23 ± 0.01a   |
| PB1                                 | 0.71 ± 0.20a    | 0.36 ± 0.23ab  | 0.31 ± 0.11ab  | 0.16 ± 0.10ab  |
| PB2                                 | 0.44 ± 0.19ab   | 0.31 ± 0.19ab  | 0.22 ± 0.09bc  | 0.14 ± 0.04ab  |
| PB3                                 | 0.25 ± 0.14b    | 0.17 ± 0.06b   | 0.13 ± 0.08c   | 0.08 ± 0.06b   |
| PC                                  | 0.35 ± 0.05a    | 0.16 ± 0.02ab  | 0.08 ± 0.05a   | 0.06 ± 0.06a   |
| PC1                                 | 0.29 ± 0.04a    | 0.24 ± 0.05a   | 0.12 ± 0.09a   | 0.02 ± 0.01a   |
| PC2                                 | 0.21 ± 0.13ab   | 0.11 ± 0.08b   | 0.12 ± 0.03a   | 0.05 ± 0.05a   |
| PC3                                 | 0.10 ± 0.09b    | 0.06 ± 0.07b   | 0.03 ± 0.01a   | 0.03 ± 0.01a   |
| Soil pesticide residues (mg/kg)     |                 |                |                |                |
| PB                                  | ND              | 0.005 ± 0.001a | 0.004 ± 0.001a | 0.005 ± 0.001a |
| PB1                                 | ND              | 0.005 ± 0.001b | 0.003 ± 0.005a | 0.001 ± 0.002b |
| PB2                                 | ND              | 0.003 ± 0.001b | 0.002 ± 0.004a | ND             |
| PB3                                 | ND              | ND             | ND             | ND             |
| PC                                  | 0.007 ± 0.004a  | 0.006 ± 0.004a | 0.012 ± 0.011a | 0.008 ± 0.004a |
| PC1                                 | 0.003 ± 0.004ab | 0.007 ± 0.010a | 0.009 ± 0.008a | 0.007 ± 0.007a |
| PC2                                 | 0.001 ± 0.002b  | 0.006 ± 0.005a | 0.007 ± 0.004a | 0.006 ± 0.007a |
| PC3                                 | ND              | 0.002 ± 0.003a | 0.003 ± 0.004a | 0.003 ± 0.006a |
| Runoff pesticide residues (mg/kg)   |                 |                |                |                |
| PB                                  | ND              | 0.002 ± 0.002a | 0.001 ± 0.001a | ND             |
| PB1                                 | ND              | 0.001 ± 0.002a | 0.001 ± 0.001a | ND             |
| PB2                                 | ND              | 0.001 ± 0.001a | ND             | ND             |
| PB3                                 | ND              | ND             | ND             | ND             |
| PC                                  | ND              | 0.001 ± 0.001a | 0.001 ± 0.001a | ND             |
| PC1                                 | ND              | ND             | ND             | ND             |
| PC2                                 | ND              | ND             | ND             | ND             |
| PC3                                 | ND              | ND             | ND             | ND             |

PB is the 100% bifenthrin pesticide treatment, PB1 is the 70% bifenthrin pesticide treatment, PB2 is the 50% bifenthrin pesticide treatment, and PB3 is the 20% bifenthrin pesticide treatment. PC is the 100% chlorfenapyr pesticide treatment, PC1 is the 70% chlorfenapyr pesticide treatment, PC2 is the 50% chlorfenapyr pesticide treatment, and PC3 is the 20% chlorfenapyr pesticide treatment. Different letters indicate significant differences ( $P \leq 0.05$ ) between different treatments according to the Student–Newman–Keuls test

ND not detected

### Comprehensive effects of different fertilizer and pesticide treatments on tea yield and quality

The yield and quality of tea are the most important factors in the process of tea cultivation. The use of fertilizer to promote the growth of tea by providing NPK nutrients to tea trees will directly affect the yield and quality of tea leaves, and pesticide application can effectively reduce the loss of tea production by preventing and killing pests. In particular, as the economic leaves are the main product, the prevention and control of tea insect pests, which will directly affect the yield and quality of tea and the income generated by tea, are crucial. For

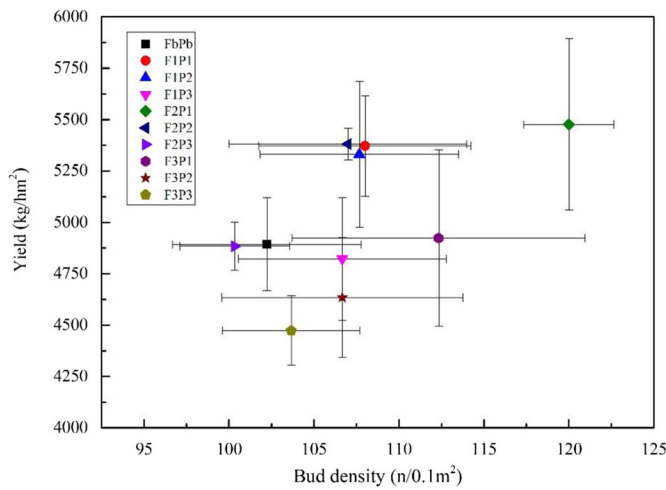
**Table 7** The degradation kinetics of tea leaf pesticide residues

| Treatment | Dynamic equation       | $R^2$         | $t_{1/2}$ /day |
|-----------|------------------------|---------------|----------------|
| PB        | $y = 0.792e^{-0.118x}$ | $R^2 = 0.982$ | 5.89           |
| PB1       | $y = 0.706e^{-0.142x}$ | $R^2 = 0.896$ | 4.87           |
| PB2       | $y = 0.482e^{-0.122x}$ | $R^2 = 0.983$ | 5.68           |
| PB3       | $y = 0.264e^{-0.113x}$ | $R^2 = 0.971$ | 6.13           |
| PC        | $y = 0.341e^{-0.187x}$ | $R^2 = 0.928$ | 3.72           |
| PC1       | $y = 0.488e^{-0.271x}$ | $R^2 = 0.886$ | 2.56           |
| PC2       | $y = 0.210e^{-0.125x}$ | $R^2 = 0.782$ | 5.55           |
| PC3       | $y = 0.096e^{-0.127x}$ | $R^2 = 0.859$ | 5.46           |

PB is the 100% bifenthrin pesticide treatment, PB1 is the 70% bifenthrin pesticide treatment, PB2 is the 50% bifenthrin pesticide treatment, and PB3 is the 20% bifenthrin pesticide treatment. PC is the 100% chlorfenapyr pesticide treatment, PC1 is the 70% chlorfenapyr pesticide treatment, PC2 is the 50% chlorfenapyr pesticide treatment, and PC3 is the 20% chlorfenapyr pesticide treatment.  $R^2$  denotes the equation fitting coefficient, and  $t_{1/2}$ /day is the pesticide half-life

average tea yield, the ten treatments followed the pattern  $F3P3 < F3P2 < F1P3 < F2P3 < FbPb < F3P1 < F1P2 < F1P1 < F2P2 < F2P1$ ; all the specific values are shown in Fig. 3. In general, the yield from the treatments combining F1 or F2 and P1 or P2 was significantly higher than that from the treatments combining Fb or F3 and Pb or P3. Regarding tea quality, better quality is generally achieved by higher polyphenol, caffeine, water extract, and amino acid contents. Especially for green tea, a typical unfermented tea, a lower tea polyphenol/amino acid (P/AA) content typically indicates a superior aroma and taste (Juneja et al. 1999; Zhang et al. 2016). The average P/AA ratio of the ten treatments followed the pattern  $F2P2 (4.71) < F2P1 (4.79) < F1P2 (4.93) < F1P3 (5.05) < F3P2 (5.06) < F2P3 (5.37) < F3P1 (5.40) < F1P1 (5.41) < F3P3 (5.45) < FbPb (5.51)$ . Organic fertilizer replacements can effectively reduce the P/AA ratio of green tea; the lowest P/AA ratio is produced when the organic fertilizer replacement is 50% and the pesticide reduction is 50%.

Multivariate ANOVA was used to further investigate the interaction effects of different fertilizer and pesticide managements on tea yield and quality ( $P \leq 0.05$ ). Based on the results, fertilizer and pesticides both had a significant effect on tea yield, but the effects of the interaction between fertilizer and pesticides on tea yield and quality were relatively weak. Further analysis was performed based on the decision coefficient  $R^2$ , which represents the proportion of variation that can be explained by the interaction between fertilizer and pesticide managements. The decision coefficients of tea yield ( $R^2 = 0.633$ ), bud density ( $R^2 = 0.522$ ), tea polyphenols ( $R^2 = 0.750$ ), and caffeine ( $R^2 = 0.669$ ) were all greater than 0.5 (Table 8). More than 50% of the variation in these four indicators can be explained by the interaction effects of fertilizer and pesticide managements. Thus, based on a comprehensive analysis of the ten treatments of the experiment, the treatments

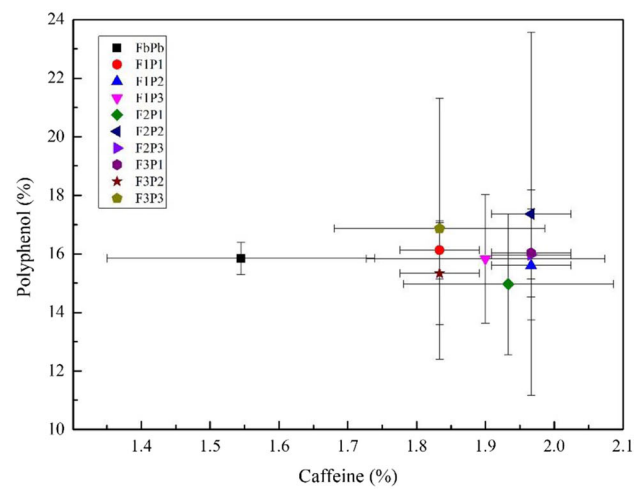


**Fig. 3** The effects of different fertilizer and pesticide managements on tea yield and quality. FbPb is the treatment of 100% chemical fertilizer and 100% pesticide; F1P1, F1P2, and F1P3 are the treatments of 20% organic fertilizer and 70% pesticide, 50% pesticide, and 20% pesticide, respectively; F2P1, F2P2, and F2P3 are the treatments of 50% organic

with 20% or 50% organic fertilizer replacement and the treatments with 20% or 50% pesticide reduction (e.g., F1P1, F1P2, F2P1, and F2P2) had the most desirable results (Fig. 3). The tea yield and quality indices of these four treatments were significantly higher than those of the other treatments. The yield in these four treatments was 4.4–17.4% higher, and the bud density was 8.9–11.9% higher than those in the FbPb treatment. The yield and bud density in treatments containing F3 and P3 were lower than those in the other treatments. The caffeine content in all other treatments was much higher than that in the FbPb treatment, i.e., increasing by 18.7–27.3% on average, though differences in tea polyphenols between treatments were not obvious.

### Discussion

This study was conducted to explore the effects of dual reductions in fertilizers and pesticides on environmental variables, including soil properties, nutrient loss from soils to runoff water, and pesticide residues in crops and environments as well as the yield and quality of tea in a tea garden. Teas are multi-harvest agricultural products (harvested multiple times each year), and fertilization and pesticide applications occur



fertilizer and 70% pesticide, 50% pesticide, and 20% pesticide, respectively; and F3P1, F3P2, and F3P3 are the treatments of 80% organic fertilizer and 70% pesticide, 50% pesticide, and 20% pesticide, respectively

several times before and after each tea harvest. In rainy areas of Southern China, heavy rains over a short period of time are the main cause of nitrogen and phosphorus loss in tea gardens. Under the influence of heavy rainfall conditions, the application of chemical fertilizers leads to the rapid and pronounced input of nutrients such as nitrogen and phosphorus in tea-garden runoff water. Therefore, it is important to understand the environmental effects of organic fertilizer replacement measures under a year-round complete fertilization cycle. Similarly, because of the rapid decay characteristics of chemical pesticides (the half-life of sprayed pesticides is only 5 days on average), short-term studies are conducive for investigating the effects of pesticide reduction on pesticide residues in tea, soil, and runoff water. As a cash crop for leaf use, only the young shoots and leaves of tea plants are harvested during the harvest seasons of spring, summer, and autumn. Our plot experiments spanned a complete cycle of fertilization replacement and pesticide reduction application over a harvest year and therefore provide a thorough understanding of the environmental effects of fertilization replacement and pesticide reduction.

The soil pH in each treatment was greater than 4.0 before fertilization and gradually decreased after fertilization. In August, the soil pH in the Fb (3.99) and F1 (3.97) treatments

**Table 8** The results of a multivariate analysis of the effects of fertilizer and pesticide managements on tea yield and quality

| Treatment                             | Yield | Bud density | Polyphenols | Caffeine | Water extracts | Amino acids |
|---------------------------------------|-------|-------------|-------------|----------|----------------|-------------|
| Significance (fertilizer)             | 0.001 | 0.796       | 0.974       | 0.479    | 0.563          | 0.601       |
| Significance (pesticide)              | 0.001 | 0.005       | 0.903       | 0.943    | 0.965          | 0.601       |
| Significance (fertilizer × pesticide) | 0.797 | 0.129       | 0.769       | 0.527    | 0.936          | 0.135       |
| R <sup>2</sup>                        | 0.633 | 0.522       | 0.750       | 0.669    | 0.107          | 0.499       |

was less than 4.00. Therefore, the soil acidification caused by fertilization was obvious. This is consistent with Han et al. (2002) who reported that more than 40% of tea plantations had a soil pH of approximately 4.0. Tea is a unique, perennial, leaf-harvested, commercial crop that prefers acidic soil and has an optimum pH between 4.0 and 5.5 (Ruan et al. 2007; Fung et al. 2010). When tea plantations are fertilized, the trees absorb more cations than anions, the root system secretes protons ( $H^+$ ), and soil acidification is caused by the  $H^+$  residue left after the absorption of nitrogen, which might be the main reason why the pH value was lower than that before fertilization in our study (Ruan et al. 2000, 2007). A similar nitrogen application field experiment conducted in a tea plantation in Zhejiang Province also found that a high nitrogen application rate can accelerate soil acidification in both topsoils and soil profiles to a depth of 200 cm (Yan et al. 2018). To effectively reduce the trend of soil acidification in tea plantations and ensure the nutrient supply of tea trees, an alternative method using organic fertilizer was proposed. In this study, appropriate organic fertilizer replacement management effectively mitigated soil acidification. Specifically, compared with the soil pH in the 100% chemical fertilizer treatment, that in the 50% organic fertilizer replacement treatment and 80% organic fertilizer replacement treatment was significantly increased by 1.7–4.8% and 2.2–6.0%, respectively. Similar observations were also reported, whereby organic fertilizer had lower nitrification potential than did inorganic nitrogen fertilizer at the same nitrogen level because the nutrient content in the organic fertilizer was lower than that in the chemical fertilizer (Heeb et al. 2010; Chu et al. 2007; Liang et al. 2012).

Clearly, replacement with organic fertilizers can provide the soil with more OM. In this experiment, the OM content of the F2 and F3 treatments was 7.0–54.7% and 4.3–39.1% higher, respectively, than that in the Fb treatment. Richer soil OM content can promote soil microbial activity and the conversion of soil nutrients, ultimately increasing the soil nutrient content. Compared to  $NO_3$ -N, the  $NH_4$ -N content in soil was significantly higher, and the  $NH_4$ -N content in the organic fertilizer replacement treatments was significantly higher than that in the Fb treatment. Therefore, for tea trees that prefer  $NH_4$ -N, organic fertilizer replacement is more conducive to the growth of tea trees than is chemical fertilizer. In addition,  $NO_3$ -N contents in runoff water were higher than were  $NH_4$ -N contents.  $NH_4$ -N is usually adsorbed by soil particles and OM (Ardakani et al. 1974), and under aerobic soil conditions,  $NH_4$ -N can change rapidly to  $NO_3$ -N (Koper et al. 2010). The nitrification activity of tea plantation soils is very high, and when fertilizer is applied, the number of nitrifying bacteria will increase, resulting in rapid conversion of  $NH_4$ -N to  $NO_3$ -N (Walker and Wickramasinghe 1979). Furthermore, with concentrated heavy rainfall in the summer in the south,  $NO_3$ -N from fertilizers and soil nutrients will quickly be incorporated into runoff water during the rainfall process. All

these factors may explain why the nitrogen in the tea plantation runoff water is mainly  $NO_3$ -N.

The Olsen P and Olsen K contents were also slightly higher in the soil of the organic fertilizer replacement treatments than in the soil of the Fb treatment, which may be due to the fact that organic fertilizers are released relatively slowly; in contrast, chemical fertilizers are released quickly and easily with the flow of water. The Olsen P and Olsen K contents in runoff water also confirmed this mechanism. Rapid loss of nutrients such as NPK is an important risk factor of agricultural non-point pollution, especially in hilly areas such as the study area, where there is heavy rainfall in summer. Previous studies also support that the amount of nitrogen and phosphorus loss caused by heavy rain after fertilization in agricultural watersheds is often the highest of the entire year (Udawatta and Motavalli 2006). In this study, the correlation between NPK and rainfall also indicated that the total amount of rainfall is the main cause of the loss of soil  $NH_4$ -N and  $NO_3$ -N; however, both total rainfall and heavy rain in a short time are important causes of potassium and phosphorus loss through runoff in experimental tea plantations.

A reduction in pesticide application is known to reduce pesticide residue in tea. In general, pesticide residues of tea decreased with an increase in pesticide reduction and time. The degradation kinetics of the bifenthrin and chlorfenapyr residues in tea leaves both followed a first-order dynamic equation, indicating rapid degradation. In fact, because of the volatilization and degradation characteristics of pesticides and the impact of rainfall, the amount of pesticide residues left on the tea leaves after spraying is very low, and the pesticide content in the soil and water bodies is minimal. Moreover, the pesticides gradually degraded after spraying, and the effect of reducing the application of pesticides to different levels decreased. Therefore, it is not necessary to reduce the use of pesticides in tea plantations below the safety level specified by the manufacturer. Increasing the number of sprays applied within the pesticide residue limits may be helpful in improving tea yield and quality.

The managements of organic fertilizer replacement and pesticide application can effectively influence tea yield and improve the quality of green tea by reducing the P/AA ratio. By analyzing the interactive effects of different fertilizer and pesticide managements, a reasonable application scheme of fertilizers and pesticides can be obtained. More than 50% of the variation in tea yield, bud density, polyphenols, and caffeine was explained by interactions between fertilizers and pesticide managements (Table 8). The results indicated that the use of organic fertilizers can increase the yield of tea, though the yield will not continue to increase when the replacement with organic fertilizers exceeds a certain proportion, such as 80%. Similarly, when pesticides are reduced to 80%, the yield was also lower than that in the treatment in which pesticides were reduced by 30%. The combinations of

organic fertilizer replacement of 20% or 50% with a pesticide application reduction of 30% or 50%, such as those used in the F1P1, F1P2, F2P1, and F2P2 treatments, are ideal combinations for balancing the yield and quality of tea. The tea yield and quality were highest when an organic fertilizer replacement of 50% and a pesticide reduction of 50% (F2P2) were used (Fig. 3). Excessive replacement with organic fertilizers and reduction in pesticides will be less effective than will 100% chemical fertilizers and 100% pesticide application, such as that used in the F3P2 and F3P3 treatments.

## Conclusions

The management of replacing conventional chemical fertilizer with organic fertilizer has the potential to improve the physical and chemical properties of tea plantation soil. Field experimental results showed that replacement with organic fertilizer can effectively elevate soil pH, buffer soil acidification, and increase soil OM, NH<sub>4</sub>-N, Olsen P, and Olsen K contents while decreasing NH<sub>4</sub>-N, NO<sub>3</sub>-N, TP, and TK inputs in tea plantation runoff. Furthermore, the reductions in runoff nutrient inputs were positively correlated with the proportions of organic fertilizer replacement. Heavy rainfall during the rainy season contributes greatly to the input of NPK nutrients in runoff in green tea plantations; therefore, management with proper proportions of organic fertilizer replacement can be conducive to both maintaining soil fertility and reducing agricultural non-point pollution. Moderate reductions in bifenthrin and chlorfenapyr doses can reduce pesticide residues in tea leaves. However, because of the rapid degradation of pesticides, the effect of pesticide residue reduction at the late stage of spraying is not significant.

The application of fertilizers and pesticides contributed greatly to the yield, bud density, polyphenols, and caffeine of tea. The combinations of organic fertilizer replacement of 20% and a pesticide application reduction of 30% (F1P1), organic fertilizer replacement of 20% and a pesticide application reduction of 50% (F1P2), and organic fertilizer replacement of 50% and a pesticide application reduction of 30% (F2P1) produced excellent results. Among all combinations, that with organic fertilizer replacement of 50% and pesticide application reduction of 50% (F2P2) was best for balancing the yield and quality of tea.

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