Effects of reducing chemical fertilizer on the quality components of Tieguanyin tea leaves

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Abstract. Fertilizer plays an important role in agricultural development. However, the soil environment of tea gardens has been deteriorated and the growth of tea trees has been hindered by the long-term excessive chemical fertilizer used in tea gardens. To study effects of reducing chemical fertilizer on the quality components of Tieguanyin tea leaves, Tieguanyin tea garden located in yellow-red soil was treated with three consecutive times and five recipes of chemical fertilizer. The results showed that appropriate reduction of chemical fertilizer applied in tea gardens can increase the contents of potassium and organic matter in soil, polyphenols and water extract in tea leaves. Most of the contents reach their peaks in T3 treatment, which compound fertilizer that the contents of NPK are all 15% was 412.50 kg/ha and urea was 206.25 kg/ha applied in the tea gardens, that 55% of the conventional usage by local farmers. At the same time, with the reducing chemical fertilizer applied in tea gardens, the caffeine in tea leaves was significantly decreased. Therefore, T3 treatment can improve the soil environment of tea gardens and the concentration of Oolong tea soup. Besides, it infers that the amount of chemical fertilizer in T3 treatment is a key control point for the quality of tea. The indirect influence coefficients that the chemical fertilizer effects on the contents of tea water extract, free amino acid and caffeine are all at their peaks of 5.249, 4.245 and 8.594 respectively through the changing of the soil nutrient contents. It is benefit for improving the overall quality of Oolong tea.

1. Introduction
Tea is one of the three major beverages in the world. By the end of 2017, the global tea garden area was 5.09 million hectares, and China accounted for 60% of it [1]. Nitrogen is one of the important nutrient elements in the growth process of tea trees (Camellia sinensis (L.) O. Ktze), which also plays a key role in improving the quality of tea leaves. Therefore, many tea farmers pursue high yield and quality of tea trees, chemical fertilizers had been excessive application in tea gardens [2]. Among them, nitrogen fertilizer (urea, ammonium carbonate, etc.) are most widely used [3]. Since the end of 20th century, unreasonable fertilizer application has seriously affected the ecological environment of tea garden, resulting in serious soil acidification, hardening, nutrient loss, uneven quality of tea leaves,
low utilization rate of fertilizer in tea garden and other prominent problems. Recently, some researches on chemical and substitute fertilizer focus on improving application methods and affective factors on grain, fruit and vegetable economic crops. The methods including chemical fertilizer combined with organic fertilizer [4, 5], straw is covered with soil in field and chemical fertilizer reduced [6], application of drip irrigation and mulch [7]. These investigations indicated that the reduction of chemical fertilizer, application of organic fertilizer, and adding crop straws can meet the nutrient requirements in different growth stages of plants [8], which conducive to up-regulated soil enzyme activity [9] and increase fungi and bacteria colonies in the soil [10]. Furthermore, it also promotes the decomposition of soil organic matter and fertilizer conversion rate, and further improves crop yield and quality. Different combinations of substrates and fertilizers have been carried out on crops such as vegetables, melons and fruits. Studies have shown that the application of different proportions of biogas slurry can improve the total chlorophyll content and yield of seaweed, and have good effects on the growth, yield and quality characteristics of ornamental lettuce. Compared with conventional fertilizers, the plant height of lettuce increased by 16.75%, the leaf area increased by 42.03%, and the contents of vitamin C, anthocyanin, soluble sugar and protein were higher than those of the control group. Reducing the application of chemical fertilizers and increasing the effective utilization rate of chemical fertilizers have become an important research content of workers in agriculture and horticulture.

However, there is weak in the research on the simple reduction of chemical fertilizers in tea gardens. The research on the path and size of the influence of chemical fertilizers on tea quality components is still unclear, and cannot be used as references for reducing chemical fertilizers and improving tea quality in tea gardens. Representative Tieguanyin tea gardens were chosen to study the effect of tea quality under different fertilizer treatment. The objective of this study was to explore the response mechanism of tea leaves quality to the reduction of chemical fertilizer application, as well as to find out the optimum chemical fertilizer application in tea garden and provides a reference for reducing chemical fertilizer application in tea garden.

2. Materials and methods

2.1. Location and materials
The experiment was conducted from March 2017 to October 2017 in Ju Yuan village, Longjuan Village, Anxi County, Quanzhou, Fujian Province, China. Tea gardens are located at 117.8111°E, 24.9543°N, the altitude is 510m and it is yellow-red soil. The soil conditions of tea garden: total nitrogen content 2.10g/kg, total phosphorus content 0.13g/kg, total potassium content 14.89g/kg, alkali nitrogen content 39.94mg/kg, available phosphorus content 4.60mg/kg, available potassium content 70.67mg/kg, organic matter content was 1.19%. The test tea tree variety is Tieguanyin that usually be made into Oolong tea, and the age of the trees is 5 years old.

Using the conventional fertilization by tea famers that urea 375kg/ha, compound fertilizer (the contents of NPK are all 15%) 750 kg/ha as the control (CK), and set the four fertilizer treatment levels, respectively: T1 - conventional fertilization * 85%, that is, urea 318.75kg/ha, compound fertilizer 637.5kg/ha; T2 - conventional fertilization * 70%, that is, urea 262.5kg/ha, compound fertilizer 525kg/ha; T3 - conventional fertilization * 55%, that is, urea 206.25kg/ha, compound fertilizer 412.5kg/ha; T4 - conventional fertilization * 40%, that is, urea 150kg/ha, compound fertilizer 300kg/ha. Each treatment level was repeated 3 times in a random arrangement, 50m² per cell. Three times of fertilization were applied on March 10 (spring fertilizer), June 1 (summer fertilizer) and July 15 (autumn fertilizer) in 2017. The method of fertilization is spreading. Soil and tea leaves samples were collected on May 10 (spring tea), July 1 (summer tea), and August 15 (autumn tea) in 2017.

2.2. Experimental methods
Soil samples collected: S-type, five-point sampling method was used in this study. That is, in each test plot, five points are selected according to the S-shaped inflection point, and the surface weeds, dead
branches and topsoil are removed, and the 0~30 cm soil layer is taken, the soil samples were thoroughly mixed and placed in the room to be air-dried for testing.

Methods for soil nutrient detection were referred to NY/T 1121.24-2012 for total nitrogen determination; LY/T 1229-1999 for alkali nitrogen determination; LY/T 1332-1999 for total phosphorus determination; NY/T 1121.7-2006 for available phosphorus determination; LY/T 1234-1999 for total potassium determination; LY/T 1236-1999 for available potassium determination; and NY/T 1121.6-2006 for organic matter determination, all of China.

Tea leaves samples collected: the plucking standard is three tea fresh leaves, each plot uses the measuring frame (0.1m²) to randomly pick 10 frames fresh leaves, and use the blast dryer under 120°C degrees for 10 minutes and then right away dried under 80°C degrees to dry. Methods for tea quality determination were referred to GB/T 8313-2008 for tea polyphenols determination (Foline-phenol colorimetric method); GB/T 8312-2013 for caffeine determination; GB/T 8305-2013 for water extracts determination; GB/T 8314-2013 for free amino acids determination, all of China.

2.3. Data processing
Data processing uses Excel.2010 and SPSS.21 statistical analysis software.

3. Results

3.1. Changes of soil nutrient contents in tea gardens treated with different fertilizer application rates

3.1.1. Changes of nitrogen content in tea garden soil treated with different fertilizer application rates. The result showed that the less fertilization amount applied, the less the total nitrogen content in the garden soil (Fig.1). The amount of decline in spring and autumn were reached a significant level, but not in summer. In addition, the lowest total nitrogen content of soil is in autumn. The reason may be the autumn fertilization is carried out in mid-July, which is the highest temperature period of the three fertilization treatments, and large amount of nitrogen fertilizer are volatilized. From the stability of N-bank, based on spring, in summer and autumn, the total nitrogen content variation in CK, T1, T2, T3 and T4 was 13.08%~20.39%, 21.88%~45.46%, 13.95%~52.35%, 0.36%~21.05%, 0.95%~39.86%, respectively. In which, the CK and T3 have the smallest variation, indicating that the soil nitrogen pool is stable under the treatment of 100% fertilization and 55% fertilization.

![Figure 1](image_url)
Alkaline nitrogen acts as a major source of nitrogen which directly acting on tea trees and plays a vital role in tea yield and quality [11]. The data (Fig. 2) shows that the content of alkali nitrogen in soil decreased with fertilization amount in tea garden, which is consistent with the change in soil total nitrogen content, indicating that the total nitrogen content in soil and alkali nitrogen content have synergistic effects. Based on the soil alkali nitrogen content in spring, the changes in soil alkali nitrogen content under CK, T1, T2, T3 and T4 fertilization treatments were 29.03%~45.38%, 1.18%~23.70%, 1.68%~13.39%, 17.22%~36.68%, 2.77%~3.16%, respectively. For the fluctuation range, the largest in CK and the smallest in T4. The change in alkali nitrogen content was correlative with fluctuation of total nitrogen content. In the same fertilization treatment, the content of alkali nitrogen in CK, T1 and T3 treatments decreased first and then increased, while T2 and T4 treatment stabilized. It indicated in T2 and T4 treatment, the conversion rate of alkali nitrogen in soil consistent with the plant absorption rate. It is helpful to know that maintain the relative stability of soil nitrogen was beneficial to improve fertilizer utilization.

3.1.2. Changes of phosphorus content in tea garden soil treated with different fertilizer application rates. Figure 3 shows the phosphorus content of the garden soil. The results show total phosphorus content of the soil are decreased first, then increase and then decrease again. The total phosphorus content shows that the highest content of CK treatment in spring and summer is 0.26 g/kg and 0.16 g/kg, respectively, and the highest in autumn is T3 treatment, 0.15 g/kg. These data indicate that the topdressing can significantly increase the total phosphorus content in the soil. Based on the total phosphorus content in spring, the changes in soil total phosphorus under CK, T1, T2, T3 and T4 fertilization treatments were 39.12%~49.59%, 44.59%~57.99%, 39.27%~64.62%, 7.88%~40.82% and 25.57% to 53.82%, respectively.

These data indicate that the topdressing can significantly increase the total phosphorus content in the soil. Based on the total phosphorus content in spring, the changes in soil total phosphorus under CK, T1, T2, T3 and T4 fertilization treatments were 39.12%~49.59%, 44.59%~57.99%, 39.27%~64.62%, 7.88%~40.82% and 25.57% to 53.82%, respectively.
The available phosphorus content in the soil can directly reflect the phosphorus supply capacity of soil. Fig. 4 shows that the soil available phosphorus content is significantly different under different treatments, and the highest value in CK and the lowest value in T4. T4 compared with CK are decreased about 61.57%, 53.09% and 47.29% in spring, summer and autumn, respectively. The percentage difference is decreasing. It indicates that the available phosphorus content in soil gradually approached CK value. With the continuous reduction of fertilization amount, the continuously decrease of soil available phosphorus content, which is consistent with the decrease of T4 in different seasons compared with CK treatment. It shows that application of chemical fertilizer can increase the soil phosphorus content in a short time, but the phosphorus fixed ability is limited in soil. Once the large amount of chemical fertilizer can exceed the fixed maximum of the soil, the phosphorus will be lost in the soil, reducing fertilizer utilization. It is suggested that the CK fertilization has exceeded the maximum fixed ability of the soil, therefore, it will decrease rapidly with time.

3.1.3. Changes of potassium content in tea garden soil treated with different fertilizer application rates. In addition to the large amount of nitrogen required for the growth of tea trees, potassium is also a key element for physiological activities such as photosynthesis and nitrogen metabolism of tea trees [12].

Based on the spring soil (Fig. 5), the total potassium changes in soil under CK, T1, T2, T3 and T4 fertilization treatments were 24.30%~52.18%, 16.02%~21.51%, 8.62%~14.98%, 13.12%~25.51% and 2.75%~30.67%, respectively. The range of variation is the least for T1 and T2 fertilization, and the maximum for CK and T4 fertilization. The excessive or less application of chemical fertilizer will greatly affect the total potassium content in the soil. Appropriate reduction of chemical fertilizer application enhances the soil's anti-interference and improves soil nutrient stability.

Figure 4. Consecutive chemical fertilizer reduction changed the content of soil available phosphorus

Figure 5. Consecutive chemical fertilizer reduction changed the content of soil total potassium
Soil available potassium is an important indicator for judging the abundance of soil potassium. Fig. 6 shows the available potassium content in soil is fluctuation under different treatments. Among them, the spring and autumn showed significant levels, and no significant difference in summer. The highest values in spring, summer and autumn are 98.84 mg/kg (T1), 79.26 mg/kg (T1) and 107.08 mg/kg (T3) respectively, which are higher than the lowest value 31.08%, 13.93% and 18.80%. The overall difference is small. This finding showed that the application of excessive fertilizer would not significantly increase the available potassium content in the soil, and might reduce the utilization of fertilizer.

3.1.4. Changes of organic matter content in tea garden soil treated with different fertilizer application rates. Soil organic matter directly affects soil water retention, fertilizer retention and gas permeability [13]. Fig. 7 shows the changes in soil organic contents, except spring tea, the soil organic contents in different fertilization treatments are increased with the decrease of fertilization amount, and the soil organic content from T1 to T2 treatment is increased higher than CK treatment.

Two reasons can explain. First, the temperature in summer and autumn is higher than that in spring, resulting to the higher activity for microbial growth and enzyme activity in spring. Higher enzyme activities accelerate the decomposition of animal and plant tissues and increase of soil organic matter. Second, in the spring and summer, the tea is lightly pruned about one month before harvesting. The pruning leaves directly cover the soil of the tea garden and, thus increases the soil organic content after decomposition.
3.2. Changes in main quality components of Tieguanyin leaves treated with different fertilizer application rates

Different fertilization treatments affect the main quality components of tea leaves. Table 1 shows that the reduction of fertilization can increase the content of tea water extract, especially spring tea and summer tea. Compared with CK treatment, the T3 treatment of spring tea and summer tea increased tea water extract 6.83% and 5.75%, respectively, and 6.29% in average, all at their peaks. In autumn tea, there are no significant difference between T3 and CK treatment. The reason may be fertilization of autumn tea is the hottest, and most of the fertilizers were volatilization in the high temperature environment, especially the nitrogen fertilizer is the most significant. In conclusion, appropriate reduced fertilization can slow down the fertilizer nutrient conversion and extend the action time, which is conducive to the growth and absorption of tea trees. Therefore, the reduction of fertilizer application will increase the content of tea water extract.

Comparing the changes of polyphenols content in tea leaves under different fertilization levels, it was found that the lower of fertilization amount, the higher tea polyphenols content. Kanazawa showed that nitrogen fertilizer affects the balance of carbon and nitrogen metabolism in tea [14], and the application of nitrogen fertilizer can reduce polyphenols content of tea leaves [15], the results with the study. Table 1 shows that the content of tea polyphenols in CK treatment is higher than that in T1 and T2 treatments. It may have two reasons: the first, CK treatment of long-term large-scale fertilizer application, soil microbes and enzyme activities were down-regulated, ammonification and nitrification are reduced after chemical fertilizer enters the soil [16], and the nitrogen source cannot be fixed. Second, the growth status of tea trees under CK treatment is better than other treatments, ventilation of tea gardens is reduced, and denitrification is enhanced [17], resulting in a large loss of nitrogen fertilizer. The experiment also showed that the T3 and T4 treatments in spring, summer and autumn increased by 2.55% and 0.87%, -0.44% and 5.72%, 3.37% and 4.27%, respectively. The averages were 1.82% and 3.62%, respectively.

Table 1 shows free amino acid content of tea, CK>T1>T3>T2>T4 is observed in summer tea and autumn tea, but it is irregular changing in spring tea. The free amino acid content of summer tea leaves and treat with T1, T2, T3 and T4 is reduced by 7.84%, 17.25%, 13.33% and 17.65% compare with CK fertilization. In the autumn, the free amino acid content of tea treat with T1, T2, T3 and T4 is reduced by 12.13%, 21.32%, 18.01% and 22.43% compare with CK fertilization. The averages are 9.99%, 19.29%, 15.67% and 20.04%, respectively. These results indicate that increase of fertilizer application increase the content of free amino acids. In conclusion, increase fertilizer application can increase the phenol/ammonia ratio, and improve the sensory quality of tea.

Table 1 shows the content of caffeine decrease with the decrease of fertilizer application rate, the trend is CK>T2>T1>T3>T4. However, there is no significantly different in each treatment in spring tea. It may be the spring tea grows slower than other seasons, and base fertilizer has not been fully absorbed in last year. The soil has a large amount of fertilizer residue, therefore it is no significant difference of caffeine content in spring tea. Compared with CK, the content of caffeine in T1, T2, T3 and T4 treatments in summer decreased approximately 16.18%, 5.07%, 14.49% and 21.01%, respectively. In autumn, the content of caffeine in T1, T2, T3 and T4 treatments compared with CK treatment were decreased 5.73%, 8.33%, 6.51% and 13.28%. The averages are 10.96%, 6.70%, 10.05% and 17.15%, respectively.

T2 treatment was 7.96%~36.59%; T3 treatment was 14.95%~22.05%; T4 treatment was 4.02%~16.95%.

Table 1 shows that with the decrease of fertilizer application rate, the trend is CK>T2>T1>T3>T4. However, there is no significantly different in each treatment in spring tea. It may be the spring tea grows slower than other seasons, and base fertilizer has not been fully absorbed in last year. The soil has a large amount of fertilizer residue, therefore it is no significant difference of caffeine content in spring tea. Compared with CK, the content of caffeine in T1, T2, T3 and T4 treatments in summer decreased approximately 16.18%, 5.07%, 14.49% and 21.01%, respectively. In autumn, the content of caffeine in T1, T2, T3 and T4 treatments compared with CK treatment were decreased 5.73%, 8.33%, 6.51% and 13.28%. The averages are 10.96%, 6.70%, 10.05% and 17.15%, respectively.
indicating that excessive fertilization is unfavorable for soil nutrient accumulation.

Among them, T3 treatment on organic matter, total phosphorus, total nitrogen and available phosphorus contents have the largest and positive value. T2 treatment on alkali nitrogen and available potassium contents have the largest and positive values. The total influence coefficient of T3 has the largest value (2.364) than other treatments. However, CK, T2 and T4 treatments are negative values, indicating that chemical fertilizer has a reverse effect. CK is the smallest value in these treatments, indicating that excessive fertilization is unfavorable for soil nutrient accumulation.

### Table 1. Consecutive chemical fertilizer reduction changed the main quality components of Tieguanyin tea leaves (%)

| Component          | Season       | Fertilization treatment | CK      | T1       | T2       | T3       | T4       |
|--------------------|--------------|-------------------------|---------|----------|----------|----------|----------|
| Water extract      | Spring tea   | 43.76 ± 0.19c           | 45.19 ± 0.12b | 42.79 ± 0.18d | 46.75 ± 0.21a | 45.27 ± 0.20b |
|                    | Summer tea   | 45.19 ± 0.10b           | 44.47 ± 0.19c | 44.65 ± 0.22c | 47.79 ± 0.21a | 46.55 ± 0.50b |
|                    | Autumn tea   | 48.62 ± 1.14a           | 43.24 ± 1.76b | 42.49 ± 0.56b | 48.23 ± 1.40a | 44.81 ± 1.03b |
| Tea polyphenols    | Spring tea   | 17.28 ± 0.05c           | 12.40 ± 0.03e | 13.64 ± 0.02d | 17.72 ± 0.05a | 17.43 ± 0.06b |
|                    | Summer tea   | 18.18 ± 0.02b           | 17.71 ± 0.06d | 17.93 ± 0.02c | 18.10 ± 0.07b | 19.22 ± 0.04a |
|                    | Autumn tea   | 21.08 ± 0.08b           | 20.17 ± 0.14c | 19.60 ± 0.11d | 21.79 ± 0.49a | 21.98 ± 0.17a |
| Free amino acid    | Spring tea   | 1.88 ± 0.02c            | 2.04 ± 0.02a | 1.37 ± 0.03d | 1.95 ± 0.02b | 1.86 ± 0.02c |
|                    | Summer tea   | 2.55 ± 0.02a            | 2.35 ± 0.02b | 2.11 ± 0.01d | 2.21 ± 0.01c | 2.10 ± 0.01d |
|                    | Autumn tea   | 2.72 ± 0.03a            | 2.39 ± 0.01b | 2.14 ± 0.08d | 2.23 ± 0.01c | 2.11 ± 0.05d |
| Caffeine           | Spring tea   | 3.14 ± 0.02b            | 3.42 ± 0.07a | 3.10 ± 0.05b | 3.17 ± 0.06b | 3.19 ± 0.02b |
|                    | Summer tea   | 4.14 ± 0.01a            | 3.47 ± 0.01c | 3.93 ± 0.02b | 3.54 ± 0.01d | 3.27 ± 0.01e |
|                    | Autumn tea   | 3.84 ± 0.02a            | 3.62 ± 0.01b | 3.52 ± 0.01d | 3.59 ± 0.01c | 3.33 ± 0.01e |

Note: Different letters in the table indicate significant differences at the 0.05 level.

### 3.3. The influence path analysis of chemical fertilizer reduction on quality components of leaves of Tieguanyin tea

Soil nutrients have accumulation effect, and appropriate feed with chemical fertilizer can increase soil fertility [18], which is closely related to tea tree growth and tea quality [19]. Path analysis can adjust the relationship of each factors which better than conventional regression analysis and correlation analysis [20]. It has been widely used in economic analysis, soil physical, chemical and genetic diversity analysis in recent years [21, 22]. In this experiment, the new branch of tree has a certain time to adapt to fertilizer applied. Therefore, it chooses tea leaves after treated with third reduction fertilizer as samples to do path analysis.

#### 3.3.1. Influence coefficient of different fertilization amount on soil nutrient content

Table 2 shows path analysis of chemical fertilizer use and soil nutrient content in tea gardens. In application of chemical fertilizer gradient reduction, the variation of the influence coefficient of fertilizer application on soil nutrient content is not consistent.

Among them, T3 treatment on organic matter, total phosphorus, total nitrogen and available phosphorus contents have the largest and positive value. T2 treatment on alkali nitrogen and available potassium contents have the largest and positive values. The total influence coefficient of T3 has the largest value (2.364) than other treatments. However, CK, T2 and T4 treatments are negative values, indicating that chemical fertilizer has a reverse effect. CK is the smallest value in these treatments, indicating that excessive fertilization is unfavorable for soil nutrient accumulation.

### Table 2. Influence coefficient of different fertilization amount on soil nutrient content after the third treatment

| Fertilization treatment | Organic matter | Total nitrogen | Alkaline nitrogen | Total phosphorus | Available phosphorus | Total potassium | Available potassium | Total influence coefficient |
|------------------------|----------------|---------------|-------------------|------------------|--------------------|-----------------|---------------------|---------------------------|
| CK                     | -1.360         | -0.801        | -0.582            | 0.493            | 0.273              | -2.494          | 0.884               | -3.587                    |
| T1                     | -1.944         | 2.323         | 2.201             | -2.489           | 0.090              | 2.264           | -1.503              | 0.942                     |
| T2                     | -1.960         | -1.030        | 2.307             | -0.769           | -2.055             | -0.181          | 0.953               | -2.735                    |
| T3                     | 0.028          | 1.641         | -0.675            | 2.981            | 2.508              | -1.586          | -2.533              | 2.364                     |
| T4                     | -1.007         | 0.015         | -1.313            | 0.560            | -1.677             | 0.354           | 0.854               | -2.214                    |
3.3.2. Influence coefficient of soil nutrient content on quality components of tea leaves under different fertilizer application rates. Table 3 shows the path analysis of soil nutrients and tea leaf quality components. When chemical fertilizer gradient reduction, there is no coincident influence between soil nutrient content and quality components of tea leaves, neither positive nor negative effects are also inconsistent.

T3 has the highest combine influence coefficient on free amino acids and caffeine, 1.973 and 1.440 respectively. In addition, CK has the highest combine influence coefficient on tea polyphenols and water extracts, which are 1.115 and 2.549, respectively. In conclusion, increasing the application of chemical fertilizer is beneficial to produce free amino acids and caffeine, and has little effect on water extracts, reducing the phenol/ammonia ration, which can improve the quality of Oolong tea.

Table 3. Influence coefficient of soil nutrients on quality components of Tieguanyin tea leaves after the third treatment

| Component | Fertilization treatment | Organic matter | Total nitrogen | Alkaline nitrogen | Total phosphorus | Available phosphorus | Total potassium | Available potassium | Total influence coefficient |
|-----------|-------------------------|----------------|---------------|-----------------|-----------------|--------------------|-----------------|----------------------|-----------------------------|
| Water extract | CK                      | 0.928          | 0.093         | 0.366           | -0.379          | -0.126             | 1.655           | 0.012                | 2.549                       |
|            | T1                      | -1.960         | -0.647        | 1.844           | 1.659           | -2.072             | 0.753           | -1.911               | -2.334                      |
|            | T2                      | 0.730          | 0.074         | -0.380          | -0.320          | 0.197              | -0.307          | 0.080                | 0.074                       |
|            | T3                      | -0.091         | 2.429         | -1.642          | 2.000           | 2.199              | -1.508          | -1.329               | 2.058                       |
|            | T4                      | 0.460          | -0.852        | 1.154           | -1.380          | 0.812              | -1.222          | -1.244               | -2.272                      |
| Tea polyphenols | CK                      | 0.010          | 0.108         | -0.034          | -0.151          | 0.233              | 0.857           | 0.092                | 1.115                       |
|            | T1                      | 0.677          | -1.172        | -0.517          | -0.350          | 0.536              | -0.478          | 0.507                | -0.797                      |
|            | T2                      | -0.465         | -0.258        | 0.313           | -1.016          | -0.327             | 0.255           | 0.174                | -1.324                      |
|            | T3                      | 0.071          | -0.369        | -0.241          | -0.219          | 0.349              | -0.108          | 0.508                | -0.009                      |
|            | T4                      | 0.190          | -0.284        | 0.129           | -0.427          | 0.440              | 0.014           | 0.300                | 0.362                       |
| Free amino acid | CK                      | 0.034          | -0.153        | -0.101          | -0.331          | -0.015             | 0.623           | -0.145               | -0.088                      |
|            | T1                      | -1.146         | -0.499        | 2.088           | 1.559           | -2.501             | 0.912           | -2.341               | -1.928                      |
|            | T2                      | -0.522         | -0.169        | 0.341           | -1.209          | -0.532             | 0.266           | 0.199                | -1.626                      |
|            | T3                      | 0.134          | 1.283         | -0.644          | -0.076          | 2.517              | -0.667          | -0.574               | 1.973                       |
|            | T4                      | 0.036          | -0.338        | 0.126           | -0.930          | 0.201              | -0.071          | 0.106                | -0.87                       |
| Caffeine   | CK                      | -0.016         | -0.507        | -0.152          | -0.072          | -0.080             | 0.526           | -0.433               | -0.734                      |
|            | T1                      | -2.280         | -1.040        | 2.260           | 1.983           | -2.212             | 0.789           | -2.315               | -2.815                      |
|            | T2                      | -0.464         | -0.331        | 0.427           | -1.119          | -1.075             | 0.108           | 0.180                | -2.274                      |
|            | T3                      | 0.029          | 2.149         | -0.785          | 0.350           | 2.074              | -1.081          | -1.296               | 1.440                       |
|            | T4                      | 0.021          | 0.016         | 0.063           | -0.597          | -0.008             | 0.256           | 0.586                | 0.337                       |

3.3.3. Indirect effect coefficient of different fertilization amount on the quality of Tieguanyin tea leaves through the influence of soil nutrient content. The nutrient elements of chemical fertilizers are transformed in soil, absorb by roots and then transport to correlative parts of tea tree to participate in the formation of tea quality components.

Therefore, it is necessary to consider the indirect effect of different fertilization amounts on the quality of Tieguanyin tea leaves through the influence of soil nutrient content. Table 4 shows the total indirect influence coefficients, the maximum value of free amino acids, caffeine and water extracts in T3 treated of Tieguanyin leaves is 5.249, 4.245 and 8.594, respectively. However, the coefficient of polyphenols is -2.002, which is in the middle of the five fertilizer application gradients. This indicates that T3 treatment can improve the water extracts of Tieguanyin tea leaves and reduce the ratio of phenol/ammonia, which is conducive to the formation of Oolong tea.
Table 4. Indirect effect coefficient of different fertilization amount on quality components of
Tieguanyin tea leaves after the third treatment

| Component | Fertilization treatment | Organic matter | Total nitrogen | Alkaline nitrogen | Total phosphorus | Available phosphorus | Total potassium | Available potassium | Total influence coefficient |
|-----------|------------------------|----------------|---------------|-------------------|-----------------|---------------------|----------------|---------------------|--------------------------|
| Water extract | CK                     | -1.262         | -0.074        | -0.213            | -0.187          | -0.034              | -1.554         | -0.128              | -5.888                   |
|            | T1                     | -3.810         | -1.503        | 4.059             | -4.129          | -0.186              | 2.065          | -3.519              | -6.738                   |
|            | T2                     | -1.431         | -0.076        | -0.877            | -0.246          | -0.405              | -0.048         | 0.190               | -3.014                   |
|            | T3                     | -0.003         | 3.986         | -1.108            | 5.962           | 5.515               | -1.058         | -1.454              | 8.594                    |
|            | T4                     | -0.465         | -0.013        | -1.515            | -0.773          | -1.362              | -0.025         | 0.091               | -5.621                   |
| Tea polyphenols | CK                     | -0.014         | -0.087        | -0.020            | -0.074          | 0.064               | -2.137         | 0.081               | -2.187                   |
|             | T1                     | -1.316         | -2.723        | -1.138            | -0.871          | 0.048               | -1.082         | -0.762              | -7.844                   |
|             | T2                     | -0.911         | -0.266        | 0.722             | -0.781          | -0.672              | -0.046         | 0.166               | -1.789                   |
|             | T3                     | 0.002          | -0.606        | -0.163            | -0.653          | 0.875               | -0.171         | -1.287              | -2.002                   |
|             | T4                     | -0.191         | -0.004        | -0.169            | -0.239          | -0.738              | 0.005          | 0.256               | -1.081                   |
| Free amino acid | CK                     | -0.046         | -0.123        | -0.059            | -0.163          | -0.004              | -1.312         | -0.383              | -2.077                   |
|               | T1                     | -2.228         | -1.159        | 4.596             | -3.880          | -0.225              | 1.786          | -3.479              | -4.351                   |
|               | T2                     | -1.023         | -0.174        | 0.787             | -0.930          | -1.093              | -0.020         | 0.172               | -2.292                   |
|               | T3                     | 0.004          | 2.105         | -0.435            | -0.227          | 6.313               | -1.714         | -3.283              | 5.249                    |
|               | T4                     | -0.036         | -0.005        | -0.165            | -0.521          | -0.337              | 0.091          | 0.500               | -0.999                   |
| Caffeine    | CK                     | -0.022         | -0.406        | -0.088            | -0.036          | -0.022              | -4.128         | 0.011               | -2.268                   |
|            | T1                     | -4.432         | -2.416        | 4.974             | -4.936          | -0.199              | 1.705          | -2.872              | -8.702                   |
|            | T2                     | -0.909         | -0.341        | 0.985             | -0.861          | -2.209              | -0.056         | 0.076               | -3.183                   |
|            | T3                     | 0.001          | 3.527         | -0.530            | 1.043           | 5.202               | -2.392         | -3.366              | 4.245                    |
|            | T4                     | -0.021         | 0.000         | -0.083            | -0.334          | -0.013              | -0.433         | -1.062              | 0.140                    |

4. Discussion

It is obvious to know that long-term excessive chemical fertilizer used in tea gardens has damaged the soil environment. It accelerates the acidification, harden and erosion of tea garden soil, destroys soil microorganisms, reduces soil fertility, and then hinders the growth of tea trees. At the same time, the volatilization of excessive fertilizers will affect the air quality of tea gardens. The loss of fertilizers will pollute the water body, which have been confirmed by many studies [23-25].

The results of this study indicated that reducing the application of chemical fertilizers in tea gardens, the nitrogen content was relatively stable under the T3 treatment (55% of the conventional usage by local farmers), while the total potassium, available potassium and organic matter content of the soil had been increasing, although the soil phosphorus content was decreased. The total potassium and available potassium content reached the highest level in the T3 treatment except in the summer. The organic matter content increased toward the T4 treatment with the times of fertilizer application reduced. Jianyun Ruan et al regarded that the contents of theaflavins in black tea, which is also benefit for Oolong tea was increased by K application and flavour compounds in brewed Oolong tea were considerably improved by K and Mg application [26]. It is a general consensus that the increase of organic matter content can improve soil structure, enrich soil microorganisms, and increase soil fertility of tea gardens. CC Chen et al found that the pH value, organic matter and available nutrition content is significantly or very significantly correlated [27].

This study found that comparing with CK, the amino acid content of Tieguanyin tea leaves had been decreased by 15.67%, but their water extracts and tea polyphenols increased by 6.29% and 1.82%, respectively in T3 treatment. According to the principle of material transformation in the tea making process, although the freshness (that is especially beneficial to the quality of green tea) of the Oolong tea made from this leaves will be reduced, the thickness of the tea soup will be increased, and the strong taste is the most important indicator of the quality of Oolong tea[28].

In addition, with the decreasing application of chemical fertilizers in tea gardens, the caffeine content of tea leaves decreased significantly. Compared with CK, the caffeine content in the leaves of Tieguanyin tea trees was decreased by 10.05% in T3 treatment, and 17.15% in T4 treatment. It is
benefit for the production of low-caffeine tea and helping those who are sensitive to caffeine but like to drink tea to improve their sleep.

From the correlation analysis of the results of this study (Table 2 to Table 4), with the decreasing application of chemical fertilizers in tea gardens, the quality components of water extracts, tea polyphenols, free amino acids, and caffeine in Tieguanyin tea leaves had been adjusted by the changed soil nutrients. Most of their indirect total impact coefficients are negative, except the contents of water extracts, free amino acids and theophylline in the T3 treatment and the content of the caffeine in the T4 treatment are, and all are the positive largest in the T3 treatment. It could be inferred that the fertilization amount of T3 treatment is a key point to regulate the quality of tea leaves, and this point is beneficial to the quality of Tieguanyin Oolong tea according to the above research. The result is similar to ZUO Tengda and NIE Wan’s research. The usage of fertilizers has a threshold for crop growth [29]. Excessive application of them will adversely affect soil and crop growth. Therefore, it shows that the proper amount of fertilizer can not only protect the soil and environment of the tea gardens, but also benefit the growth of tea trees and the quality of tea.

5. Conclusion

From this study, it concludes that: appropriate reduction of chemical fertilizer applied in tea gardens can increase the contents of potassium and organic matter in soil, which can improve the soil environment of tea gardens; appropriate reduction of chemical fertilizer applied in tea gardens can increase the contents of polyphenols and water extract in tea leaves, which can improve the concentration of Oolong tea soup; appropriate reduction of chemical fertilizer applied in tea gardens can decrease the content of caffeine in tea leaves, which benefits for the produce of low-caffeine tea. The amount of compound fertilizer (the contents of NPK are all 15%) 412.50 kg/ha and urea 206.25 kg/ha applied in the tea gardens (all 55% of the conventional usage by local tea farmers) is a key control point for the quality of tea leaves, it is benefit for improving the overall quality of Oolong tea. Due to the time limited and uncontrollable factors in the field, the soil nutrients of the tea garden were not stabilized, the work on the effects of soil enzyme activity and microbes after reducing fertilizer application has not been completed, and the soil characterization still not obvious. In the future, it can be conducted follow-up experiments for many years to discover the common rules for decrease chemical fertilization in tea gardens.

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References

[1] China Tea Marketing Association. The summary report of economic situation about Chinese tea industry in 2017. Available at: http://www.ctma.com.cn/ctma_news/dt/2018/0204/58999.html, Accessed on: 02/04/2018
[2] Hu X D, Geng Y B, Liang T 2018 Soil Fertilizer Sci. Chin. 1 1.
[3] Owuor P O, Gone F O, Onchiri D B, Jumba I O 1990 Food Chem. 35 59.
[4] Vaillancourt M, Chantigny M, Pageau M and Vanasse A 2018 Canadian J. Plant Sci. 98 2.
[5] Pincus L, Margenot A, Six J and Scow K 2016 Agric. Ecosystems Environ. 225 62.
[6] Li C X, Zhao F, Rui W Y, Huang, Q R, Yu, X C and Zhang W J 2009 Acta Prataculturae Sinica 18 142.
[7] Yang K J, Wang F X, Shock C C., et al. 2016 Agric. Water Manage. 4 260.
[8] Zhao Y C, Wang P, Li J, et al. 2009 European J. Agronomy 31 36.
[9] Elzobair K A, Stromberger ME, Ippolito JA, et al. 2015 Chemosphere 14 145.
[10] Muhammad K, Danial H, Muhammad B, et al. 2017 Botanical Studies 58 35.
[11] Roberts T L, Norman R J, Nathan A. Slaton, et al. 2009 Soil Science Society of Am. J. 73 2151.
[12] Jin S H, Huang J Q, Li X Q, et al. 2011 *Tree Physiology* **31** 1142.
[13] Wei W L, Yan Y, Cao J, et al. 2016 *Agric. Ecosys. Environ.* **225** 86.
[14] Kanazawa S, An G H, Kaizu T 2010 *Soil Sci. Plant Nutr.* **51** 675.
[15] Li H L, Wang L Y, Cheng H, et al. 2017 *J. Tea Sci.* **37** 383.
[16] Soares J R, Cantarella H and Menegale M L C 2012 *Soil Bio. Biochem.* **52** 82.
[17] Kakiuchi S and Misumi M 2003 *Kyushu Agric. Res.* **65** 85.
[18] Jawaria S, Ghulam A S, Khurram S, et al. 2017 *Sci. Total Environ.* **607–608** 715.
[19] YANG G R, WANG X Q, XIE J, et al. 2015 *J. Tea Sci.* **6** 574.
[20] Rani C I, Veeraragavathatham D, Sanjutha S. 2008 *J. Applied Sci. Res.* **4** 287.
[21] Li M and Wu C Y 2007 *Chin. J. Soil Sci.* **38** 43.
[22] Wei Y S, Ding S R, Zhou X C, et al. 2014 *J. Plant Genetic Res.* **15** 1364.
[23] Oh K, Kato T, Li Z P, et al. 2006 *Pedosphere* **16** 770
[24] Joydev D and Misra A K. 2010 *Int. J. Applied Environ. Sci.* **5** 11
[25] Kong L C, Cao C F, Wang Z S, et al. 2004 *Chin. J. Eco-Agric.* **12** 102.
[26] Ruan J Y, Wu X, Härder R. 1999 *J. Sci. Food Agric.* **79** 47
[27] Chen CC, Xiao B, Yu Y B, et al. 2009 *J. Northwest A. F. University (Natural Sci. Ed.),* **37** 182.
[28] Wang K B, Liu F, Liu Z H, et al. 2010 *Int. J. Food Sci. Tech.* **45** 913.
[29] Zuo T D and Nie W 2018 *Asian Agric. Res.* **10** 1.