

Article

Study on the Effect of pH on Rhizosphere Soil Fertility and the Aroma Quality of Tea Trees and Their Interactions

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Abstract: In order to fully comprehend the impact of soil acidification on the quality of tea, further analyses are essential and are of the utmost importance to the cultivation of tea trees and the simultaneous enhancement of tea quality. In May 2022, Tieguanyin tea trees planted in soils with different pH levels were selected as the research object of this study to analyze the effect of soil pH on the soil chemical index, soil fertility and the aroma quality of tea leaves. The results showed that the organic matter content, cation exchange capacity and the available nitrogen, available phosphorus and available potassium contents in the rhizosphere soil of the tea trees decreased significantly with decreasing soil pH levels (5.32–3.29), while the total nitrogen, total phosphorus and total potassium contents did not change significantly. The results of an aroma quality analysis showed that the aroma of the Tieguanyin tea was mainly floral, and the formation of floral odor characteristics was mainly derived from geraniol. The results of an interaction network analysis showed that the soil chemical indexes were significantly positively correlated with geraniol and floral aromas except for the total phosphorus and total potassium contents. In conclusion, with a decrease in the pH of soil, the soil's cation exchange capacity, organic matter content and available nutrient content showed decreasing trends which, in turn, hindered the synthesis of geraniol and reduced the floral odor characteristics of tea leaves.

Keywords: Tieguanyin (*Camellia sinensis*); soil acidification; volatile compounds; aroma intensity; odor characteristics



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1. Introduction

The Tieguanyin tea tree, also known as *Camellia sinensis*, is a perennial evergreen plant that originated in Anxi County, Fujian Province, China. The tea tree is an acidophilic plant. A soil pH of 4.5~5.5 is suitable for its cultivation, and a soil pH of less than 4.5 is unsuitable [1,2]. Lin et al. [3] analyzed the pH of soil and its effects on the yield and quality of tea at 145 tea plantations in Nanjing County, Fujian Province, and found that 82.1% of the tea plantations had soil pH levels < 4.5 and that the soil pH was significantly and positively correlated with the yield and quality of the tea. Wang et al. [4] found that 37.67% of tea plantation soils in Anxi County, Fujian Province, China, also experienced soil acidification (pH < 4.5). The yield and quality of tea showed decreasing trends after soil acidification [5]. It was evident that soil acidification reduced the yield and quality of tea and limited the development of the tea industry.

Yield and quality guarantee the economic benefits of tea plantations; in particular, the improvement of tea quality has a more important economic value [6,7]. Tieguanyin tea is a typical representative of oolong tea and is one of the top ten most famous teas in China [8]. According to the current standards for evaluating tea quality in China, the aroma index

accounts for 35% of the score in the quality evaluation of oolong tea, which is an important index for oolong tea [9]. The aroma of tea is mainly formed by the volatile compounds contained in the tea leaves [10]. During research, the content of volatile compounds in tea leaves is often used to evaluate the aroma quality of tea leaves [11]. However, there is a wide variety of volatile compounds in tea leaves, and more than 300 volatile compounds have been reported to be involved in the formation of the aroma of oolong tea [12]. The content of volatile compounds is related to the intensity of the aroma and the odor characteristics of tea, but the aroma thresholds of different compounds vary greatly, so the content of a compound is not equivalent to its contribution to the intensity of the aroma and the odor characteristics of a tea [13]. The aroma contribution of a compound depends mainly on its odor activity value (OAV), and only when the OAV value of a compound is greater than 1 may the compound actually contribute to the aroma [14]; a compound making an actual contribution will exhibit a characteristic odor [15]. Therefore, the OAV value is used to analyze changes in the aroma of tea which can reflect the effects of environmental changes on the quality of the tea's aroma more comprehensively.

It has been widely reported that soil acidification is highly susceptible to the degradation of tea quality [16–18]. Secondly, soil acidification also causes changes in the fertility of soil, especially in rhizosphere soil, which affects the uptake of nutrients by plants, leading to a decrease in plant yield or quality [19]. This change may be due to the acidification of soil altering the amount and type of plant root secretions which, in turn, affects the colonization of the rhizosphere soil by microorganisms and the soil enzyme activities, especially those related to nutrient cycling, altering the nutrient contents of a plant's rhizosphere soil and affecting plant yield and quality [20,21]. However, there are few reports on the effects of soil acidification on the fertility of rhizosphere soil and the quality of tea aroma, especially the effect of the soil fertility index on the aroma of tea evaluated using the OAV value method.

In this study, rhizosphere soil of tea trees planted on soil with different pH values was collected; in addition, one bud and three leaves were collected from each tea trees. The basic chemical indexes of the tea's rhizosphere soil and the content of volatile compounds in the tea were further determined, and the effects of soil acidification on rhizosphere soil fertility and tea aroma quality were analyzed to lay a foundation for the restoration of acidified soil and tea quality improvement in tea plantations.

2. Materials and Methods

2.1. Test Tea Plantation and Sample Collection

The origin of the Tieguanyin tea plant is in Anxi County, Quanzhou City, Fujian Province, China. The county is located at a longitude of 117°36'–118°17' E and a latitude of 24°50'–25°26' N, with an average elevation of 600 m above sea level, an average annual relative humidity of 80%, an average annual temperature of 18 °C and an average annual rainfall of 1800 mm. Based on our previous study [4,5], we chose the Tieguanyin tea plantation in Longjuan town, Anxi County, Quanzhou City, Fujian Province, China (at a longitude of 117°93' E and a latitude of 24°97' N), for our experimental study.

The tea plantations selected for this study were planted with the Tieguanyin tea tree variety, which is 7 years old. The average pH levels of the soils at the P1, P2, P3 and P4 experimental tea plantations were 3.29, 4.13, 4.74 and 5.32, covering areas of about 0.52, 0.48, 0.65 and 0.37 ha. Three replicates of 200 m² each were set up at each experimental tea plantation. In May 2022, the tea trees' rhizosphere soil and tea samples were collected. The tea plantations were not treated with fertilizer in the year of the sample collection. Ten tea trees were randomly selected, and fallen leaves on the surfaces of the tea trees were removed, the root systems of the tea trees were gently dug out, and the soil attached to the surfaces of the trees' root systems was shaken off and mixed to make one portion of tea tree rhizosphere soil, with three replications for each experimental tea plantation. Ten tea plants were randomly selected, and one bud and two leaves were collected, mixed and immediately placed in liquid nitrogen, i.e., there was one tea leaf sample with three replicates for each experimental tea plantation.

2.2. Soil Chemical Index Determination and Fertility Evaluation

This study determined the cation exchange capacity (CEC) and organic matter (OM), total nitrogen (TN), total phosphorus (TP), total potassium (TK), available nitrogen (AN), available phosphorus (AP) and available potassium (AK) contents of the rhizosphere soil. The indexes were determined using the method proposed by Wang et al. [9], in which the OM content was determined via the oxidation of potassium dichromate and external heating, the CEC was determined using neutral ammonium acetate, the TN content was determined via the Kjeldahl nitrogen determination method, the TP content was determined via alkali fusion molybdenum–antimony anti-colorimetry, the TK content was determined using a NaOH fusion flame photometer, the AN content was determined via alkali hydrolysis–diffusion, the AP content was determined via the NaHCO₃ solution–molybdenum–antimony anti-colorimetric method and the AK content was determined via the flame photometer method with ammonium acetate. The soil fertility evaluation was based on the soil fertility evaluation indexes for tea plantations listed in the current standards in the (Table S1) Environmental Requirement for Tea Plantation Area of the Ministry of Agriculture and Rural Affairs of the People’s Republic of China [22]. In this study, the results of the measured indexes were converted into fertility classes for an evaluation of soil fertility.

2.3. Extraction, Enrichment and GC-MS Analysis of the Volatile Compounds in Tea

Headspace solid-phase microextraction (HS-SPME)–gas chromatography–mass spectrometry (GC-MS) was used to extract, enrich and characterize the volatile components in tea. The collected tea samples were ground with liquid nitrogen and quickly passed through a 60-mesh (0.250 mm) filter to collect the powder. Then, 2 g of the powder was added to a 20 mL headspace flask, sealed and heated in a 50 °C hot water bath for 45 min in order to allow the volatile compounds in the tea to fully evaporate. Meanwhile, an SPME needle (50/30 µm, DVB/CAR/PDMS, Bellefonte, PA, USA) which had been pre-aged for 1 h was inserted into the headspace flask for absorption for 45 min. It was then immediately withdrawn and inserted into the GC-MS inlet for desorption for 5 min. Three replicates were conducted for each sample.

The GC-MS analysis was conducted utilizing an Agilent Technologies 7890A-5975C instrument (Agilent, Palo Alto, CA, USA) equipped with a DB-WAXETR column (60 m × 0.32 mm × 0.25 µm), using helium (purity > 99.999%) as the carrier gas at a flow rate of 1 mL/min. The temperature program commenced at 35 °C and was maintained for 5 min before gradually it was gradually increased to 150 °C at 3 °C/min, followed by a 10 °C/min increase to 240 °C and a subsequent 2 min hold. The ionization energy in EI mode was 70 eV, and the emission current was 34.6 µA. The ion source temperature was set at 230 °C, while the quadrupole and interface temperatures were set at 150 °C and 240 °C, respectively. The mass scan range was 45–500 µm.

LGC Standards (Charleston, SC, USA) provides a product named o2si smart solution A which is a mixed standard solution of n-alkanes ranging from C10 to C20 and consisting of even-numbered carbon atoms. This solution was used to measure the concentrations of the n-alkane samples as per the previously established method [23]. Utilizing the external standard method, the results from the n-alkane samples and the mixed standard solution were compared and converted to determine their concentrations.

Upon the completion of the sample testing process, volatile compounds were identified through a comparison with mass spectra from the National Institute of Standards and Technology Mass Spectrometry Library (NIST 8.0) in combination with retention indexes, similarity matches and the relative abundances of fragment ions. The concentrations of the compounds in the samples were determined by comparing them with the known concentrations of n-alkane standard solutions.

2.4. Odor Activity Value (OAV) Calculation and Odor Characteristic Analysis of Compounds

The aroma thresholds (Ts) of the volatile compounds were obtained by referring to studies of Lin [24] and Van [25,26]. To calculate the OAV value of each compound, the threshold value was used (Table S2) as follows: $OAV_i = C_i/T_i$, where OAV_i represents the OAV value of a compound i , C_i is the content of the compound i ($\mu\text{g}/\text{kg}$) and T_i represents the aroma threshold of the compound i ($\mu\text{g}/\text{kg}$). If the calculated OAV_i is greater than 1, the compound is considered an odor-active compound and can be used for further analyses. The total OAV value (OAV_t) represents the intensity of the tea's aroma, which is calculated by adding up the OAV_i values of the odor-active compounds ($OAV_t = \sum OAV_i$).

According to Table S2, aroma compounds with $OAV \geq 1$ can be categorized into six different odor characteristics: woody, floral, burnt, green, fruity and fatty [24]. The OAV values of the six odor characteristics were converted based on the OAV values of the compounds and their index I(I) in each odor characteristic.

2.5. Statistical Analysis

The experimental data were presented as means \pm standard error (SE). The t -test was performed using SPSS Statistics 19 software to analyze the statistical significance of the differences between the two groups. A heat map was created using Heml 1.0 software. A relational network plot was generated using Gephi 0.9.2 software. Raincloud and Principal component plots were produced using RStudio 4.2.3 software. A Venn diagram was produced using an online website at <http://jvenn.toulouse.inra.fr/app/example.html> (accessed on 12 April 2023).

3. Results and Discussion

3.1. Effect of Soil pH on Soil Fertility

Plants need a suitable environment for normal growth, and the nutrient status of the soil is one of the most important factors. Soil nutrient levels directly affect the yield or quality of plants [27]. In this study, the chemical indexes of the rhizosphere soil of tea trees planted in soils with different pH levels were analyzed. The results showed (Table 1) that the contents of the soil CEC and OM, AN, AP, and AK increased significantly with an increasing soil pH (3.29–5.32), as shown by increases from 7.26 to 22.48 cmol/kg, 8.32 to 17.96 g/kg, from 35.23 to 91.38 mg/kg, from 3.13 to 15.12 mg/kg and from 49.25 to 124.95 mg/kg, respectively, while the soil contents TN, TP and TK did not change significantly. The soil chemical indexes are closely related to the soil fertility grade and can be used to analyze the fertility status of the soil [28]. According to the grade evaluation standard of the environmental fertility of tea planting soil (Table S2), the results of this study found that when the soil pH was 5.32 and reached grade I, all the chemical indexes measured in the soil reached grade I except for the TP and TN contents, which were grade II; when the soil pH was 4.74 and reached grade II, the contents of TN, TP and AP in the soil reached grade I, and the other indexes were grade II; when the soil pH was 4.13 or 3.29 and was grade III, the contents of TN and TP in soil still reached grade I, the TK content was grade II and the other indexes were grade III.

The soil nutrient content and availability are reflections of the soil's fertilizer supply capacity, especially the available nutrients, and affect the nutrient uptake capacity of plants [29]. The pH of soil has little effect on the change in the total nutrient content in the soil, but it has a great effect on the content of available nutrients, and a high content of available nutrients is conducive to the growth of plants and improves their yield [19]. In addition, it has been reported that an increase in soil pH, most importantly, enhances the multiplication of nutrient-cycling-related microorganisms and enzyme activities in the soil, thereby promoting nutrient cycling in the soil and increasing the available nutrient content in the soil which, in turn, promotes the growth of tea trees [30,31]. It can be seen that with an increase in the soil pH, the contents of TN, TP and TK in the tea rhizosphere soil did not change significantly, but the soil CEC and the contents of OM, AN, AP and AK increased significantly with higher levels of soil fertility.

Table 1. Basic chemical indexes of different soil samples.

Index	Measured Values of Different Indexes				Fertility Levels of Different Indexes			
	P1	P2	P3	P4	P1	P2	P3	P4
pH value	3.29 ± 0.13d	4.13 ± 0.17c	4.74 ± 0.12b	5.32 ± 0.09a	III	III	II	I
Organic matter (g/kg)	8.32 ± 0.24d	9.57 ± 0.18c	12.08 ± 0.26b	17.96 ± 0.53a	III	III	II	I
Cation exchange capacity (cmol/kg)	7.26 ± 1.16d	13.52 ± 0.89c	17.13 ± 1.21b	22.48 ± 1.25a	III	III	II	I
Total nitrogen (g/kg)	2.46 ± 0.12a	2.48 ± 0.13a	2.53 ± 0.09a	2.49 ± 0.14a	I	I	I	I
Total phosphorus (g/kg)	1.24 ± 0.13a	1.09 ± 0.11a	1.15 ± 0.08a	1.13 ± 0.09a	I	I	I	I
Total potassium (g/kg)	6.84 ± 0.39a	6.95 ± 0.54a	7.42 ± 0.43a	7.38 ± 0.65a	II	II	II	II
Available nitrogen (mg/kg)	35.23 ± 1.87d	46.12 ± 2.46c	84.95 ± 1.74b	91.38 ± 2.86a	III	III	II	II
Available phosphorus (mg/kg)	3.13 ± 0.22d	4.06 ± 0.23c	10.41 ± 0.46b	15.12 ± 0.77a	III	III	I	I
Available potassium (mg/kg)	49.25 ± 2.13d	66.18 ± 2.45c	113.24 ± 2.94b	127.95 ± 2.75a	III	III	II	I

Note: I—the soil has good fertility; II—the soil's fertility is moderate; III—the soil's fertility is poor; Different lowercase letters indicate a significant difference at the $p < 0.05$ level among different samples.

3.2. Effect of Soil pH on Volatile Compounds in Tea Leaves

The results of the analysis of the effect of soils with different pH values on the volatile compounds in tea leaves (Figure 1A, Table S3) showed that soil pH could significantly affect the amount and content of volatile compounds in tea leaves. When the soil pH was 3.29, 52 volatile compounds were detected in tea leaves at 22.966 $\mu\text{g}/\text{kg}$. When the soil pH was 4.13, 50 volatile compounds were detected at 31.142 $\mu\text{g}/\text{kg}$. When the soil pH was 4.74, 69 volatile compounds were detected at 46.238 $\mu\text{g}/\text{kg}$. When the soil pH was 5.32, 73 volatile compounds were detected at 60.084 $\mu\text{g}/\text{kg}$. A further analysis showed that the same 36 volatile compounds were detected in tea leaves grown in soils with different pH levels. Secondly, when the soil pH levels were 3.29, 4.13, 4.74 and 5.32, there were 7, 0, 2 and 11 compounds unique to the tea leaves, respectively (Figure 1B). It has been reported that when plants were subjected to external environmental stress, short-term stress stimulated the synthesis and accumulation of volatile substances in plants, while long-term stress was detrimental to plant growth and reduced the synthesis and content of volatile substances in plants [32–35]. It is evident that acidification of tea-tree-planting soils has long affected the growth of tea trees, leading to decreases in the type and content of volatile compounds in tea leaves which, in turn, has affected the quality of tea leaves.

A further analysis showed (Figure 1C) that the volatile compounds of the tea tree leaves could be mainly divided into seven categories, namely terpenoids, hydrocarbons, heterocycles, alcohols, ketones, aldehydes and esters, and the volatile compounds behaved most similarly to tea trees planted in soil with a pH of 5.32 and in soil with a pH of 4.74, followed by soils with pH levels of 4.13 and 3.29. Tea trees planted in soils with different pH levels had the highest contents of terpenoids in their leaves, with terpenoids accounting for 52.83% (pH 3.29), 65.98% (pH 4.13), 63.70% (pH 4.74) and 65.60% (pH 5.32) of the total volatile compounds, respectively. Das et al. [36] studied the terpenoid contents in the leaves of different tea tree varieties and found that terpenoids accounted for the highest proportion of volatile compounds in tea leaves, and these terpenoids played an important role in the formation of tea aroma. Yin et al. [37] also found that the main aroma-contributing compounds in tea leaves were terpenoids when they studied the composition of tea aroma compounds. Many scholars have also concluded that aroma intensity was significantly related to the terpenoid contents in tea leaves and that the higher the content, the higher the quality [38–40]. Wang et al. [41] found that the expression of 7 out of 10 terpenoid-synthesizing genes in tea leaves showed a trend of increasing with an increase in the soil pH. This suggests that the increase in the soil pH was favorable to the synthesis of terpenoids in tea leaves. In summary, the quantity and content of different types of volatile compounds in tea leaves changed significantly with changes in the soil pH, with the most obvious change occurring in the contents of terpenoids. Moreover, the terpenoid contents decreased significantly with the decrease in soil pH which, in turn, led to a decrease in the aroma intensity of the tea leaves.

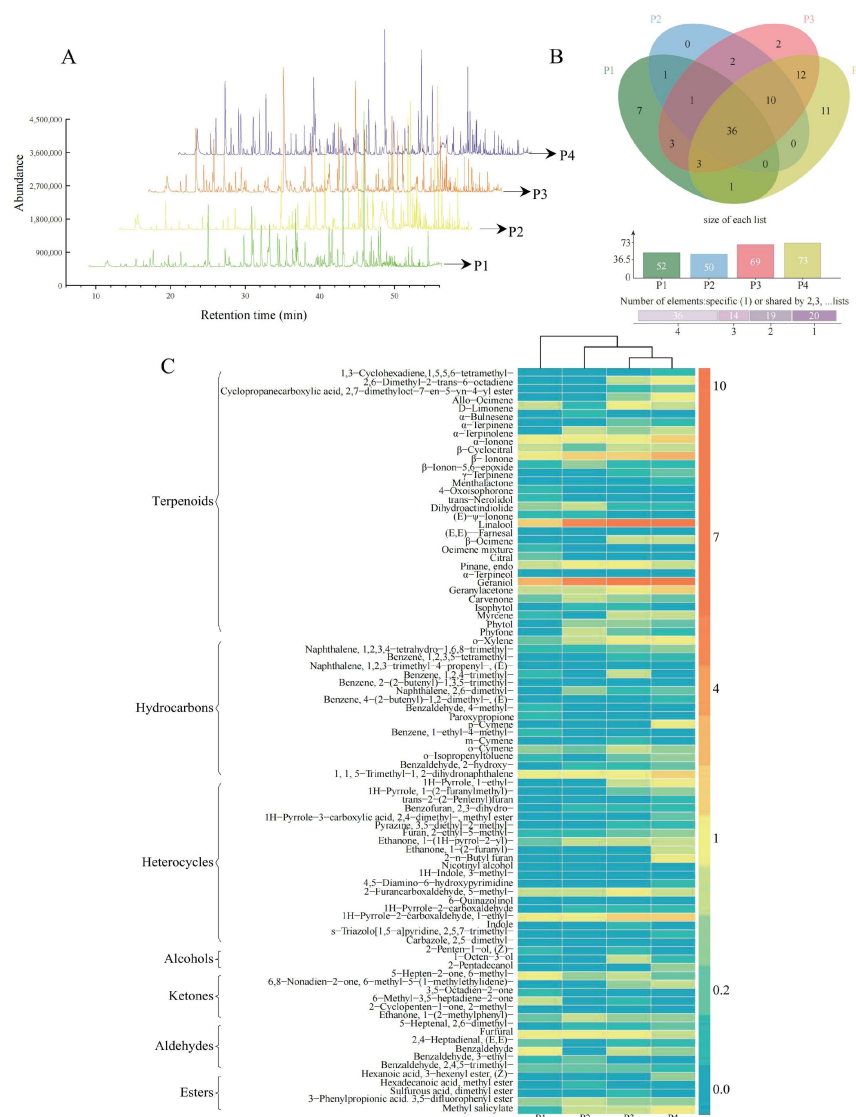


Figure 1. Effects of soil pH on volatile compounds in tea leaves. Note: P1—soil pH of 3.29; P2—soil pH of 4.13; P3—soil pH of 4.74; P4—soil pH of 5.32. (A) GC-MS map of volatile compounds in tea tree leaves; (B) Venn diagram analysis of volatile compounds in tea tree leaves; (C) Analysis of volatile compound contents in tea tree leaves.

3.3. Effect of Soil pH on Odor Activity Value (OAV) of Key Compounds in Tea Leaves

The aroma of a compound is closely related to its OAV value. When the OAV value of a compound is >1, it indicates that the compound makes an actual contribution to the aroma, and the higher the OAV value, the greater the contribution is [14,42]. Odor characteristics are analyzed and presented via compounds that contribute to the tea aroma in the aroma formation process [15,43]. In-depth analyses of the OAV values and odor characteristics of compounds are important for revealing the effect of soil pH on the formation of tea aroma. The results of this study showed (Tables S4–S7) that the contents of key compounds in tea leaves grown in soils with different pH levels were 11.984 µg/kg at a pH of 3.29, accounting for 52.18% of the total compound content; 17.935 µg/kg at a pH of 4.13, accounting for 57.95% of the total compound content; 28.228 µg/kg at a pH of 4.74, accounting for 61.05% of the total compound content; and 34.410 µg/kg at a pH of 5.32, accounting for 57.27% of the total compound content. Furthermore, when the soil pH was 3.29, 12 compounds had OAV values greater than 1, with a total OAV value of 1611.45, among which the top three contributing compounds were geraniol (65.66%), α-ionone (10.60%) and β-ionone (10.27%),

and their total contribution rate was 86.53% (Figure 2A, Table S4). When the soil pH was 4.13, 11 compounds had OAV values greater than 1, and the total OAV value was 2342.90, among which the top three contributing compounds were geraniol (70.80%), β -ionone (12.21%) and α -ionone (9.70%), and their total contribution rate was 92.71% (Figure 2A, Table S5). When the soil pH was 4.74, 12 compounds had OAV values greater than 1, and the total OAV value was 3445.46, among which the top three contributing compounds were geraniol (74.97%), β -ionone (8.52%) and α -ionone (7.72%), and their total contribution rate reached 91.21% (Figure 2A, Table S6). When the soil pH was 5.32, 12 compounds had OAV values greater than 1, and the total OAV value was 4563.82, among which the top three contributing compounds were geraniol (77.18%), β -ionone (9.03%) and α -ionone (6.83%), and their total contribution rate reached 93.04% (Figure 2A, Table S7).

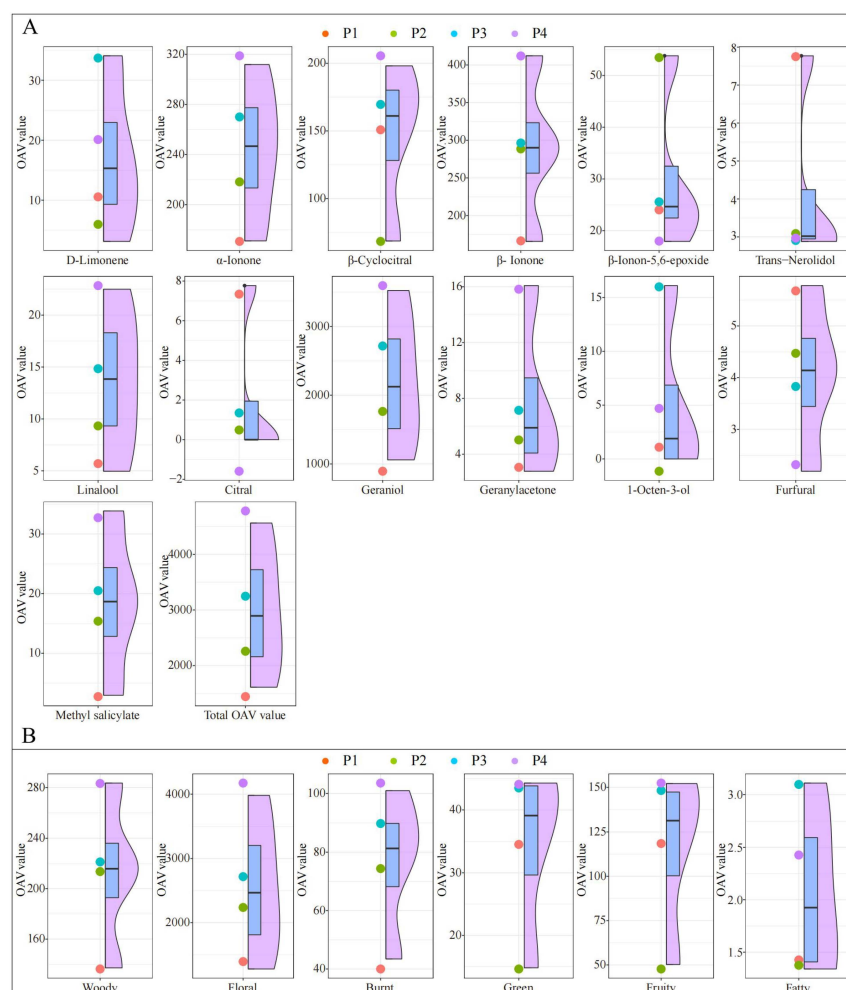


Figure 2. Analysis of odor activity values of key compounds and their odor characteristics of tea leaves in soils with different pH. Note: P1: Soil pH 3.29; P2: Soil pH 4.13; P3: Soil pH 4.74; P4: Soil pH 5.32; (A) Analysis of OAV values of key compounds; (B) OAV value analysis of odor characteristics of key compounds.

It can be seen that with the decrease in soil pH (5.32 to 3.29), the total OAV value of the aroma compounds in the tea leaves showed a downward trend (4563.82 to 1611.45), and the aroma of the tea leaves decreased. Secondly, it was found that geraniol, β -ionone and α -ionone were the major compounds contributing to the aroma of the tea leaves, showing that the total contribution rate of the three compounds to the aroma was more than 86.53%. Many scholars also found that geraniol, β -ionone and α -ionone were the main components of the aroma characteristics of tea leaves, especially geraniol [44,45]. Xiao et al. [46] found that the aroma of yellow tea was mainly formed by six compounds with OAV values greater

than 1, among which geraniol and β -ionone contributed the most. Huang et al. [47] found that geraniol was one of the key compounds in the aroma formation of black tea, and its content determined the intensity of the aroma of black tea. Niu et al. [48] analyzed the degrees of contribution of the main aroma compounds to the intensity of the aroma of different black teas and found that geraniol was the compound that made the greatest contribution in black tea. It can be seen that the formation of the aroma characteristics of tea leaves is closely related to the geraniol content in the tea leaves, and the key to the decrease in the aroma quality of the Tieguanyin tea leaves is soil acidification, as the acidification caused a decrease in the geraniol content in the tea leaves.

The odor characteristics of the compounds in tea can be mainly divided into six types, namely woody, floral, burnt, green, fruity and fatty [24]. Based on the determination of the compound contents and with reference to the contribution of different compounds to different odor characteristics (Table S1), this study further divided the odor characteristics of the key compounds into six categories, namely woody, floral, burnt, green, fruity and fatty. An analysis of the effect of soil pH on odor characteristics showed (Figure 2B) that with an increase in the soil pH (3.29~5.32), the OAV values of the woody, floral and burnt odor characteristics of the tea leaves showed a significant increase, while the OAV values of the green and fruity characteristics decreased and then increased, and the OAV value of the fatty characteristic showed a change in volatility. A further analysis revealed that the floral OAV value the floral characteristic was the largest among the different odor characteristics of tea leaves, and the floral odor presented a significant trend of increasing with an increase in the soil pH. Secondly, this study also found that the floral odor characteristics of the Tieguanyin tea mainly came from the contribution of geraniol compounds (Tables S3–S6). It has been reported that geraniol is a monoterpene, and in plant biosynthesis, lauricene is the raw material for the synthesis of geraniol, whereas when the soil pH decreases, the levels of expression of monoterpene synthase genes and lauricene synthase genes in tea tree leaves show a decrease which, in turn, decreases the geraniol content [41]. Furthermore, a floral fragrance is the main aroma characteristic of Tieguanyin tea, and the stronger the aroma, the better the aroma quality of the tea and vice versa [49–51]. It can be seen that soil acidification caused a decrease in the aroma quality of Tieguanyin tea, mainly due to the decrease in the geraniol content in the tea leaves which, in turn, led to a decrease in the strength of the floral odor characteristics.

3.4. Principal Component and Interaction Network Analysis of Different Indexes

The results of a principal component analysis based on soil chemical indexes, key aroma compounds and their odor characteristics showed (Figure 3) that different indexes could be divided into two principal components, and principal component 1 had the largest contribution rate, ranging from 66.5% to 70.2%, and could effectively distinguish different samples. Secondly, it was found that P1 (soil pH of 3.29) and P2 (soil pH of 4.13) were acidified soils and were mainly distributed at the negative end of principal component 1, while P3 (soil pH of 4.74) and P4 (soil pH of 5.32) were suitable soils for tea tree planting and were mainly distributed at the positive end of principal component 1. A further analysis revealed that all the soil chemical indexes except TP were distributed at the positive end of principal component 1, and eight key compounds and six odor characteristics were distributed at the positive end of principal component 1. The above indexes played a major role in distinguishing acidified soils from non-acidified soils. The results of the interaction network analysis between pH and the chemical indexes of the tea rhizosphere soil showed (Figure 4) that pH was significantly and positively correlated with the CEC and the AN and AK contents; the OM content was significantly and positively correlated with the AP content; the CEC was significantly and positively correlated with the AK content; and there was a significant and positive correlation between the AN, AK and AP contents. It was reported that when the soil pH was below 8.0, the CEC showed an upward trend with the increase in the soil pH, and the larger the CEC, the more beneficial the decomposition of the soil organic matter, the more beneficial the increase in the available nutrient content in the

soil and the greater the improvement in the soil's fertility [52–54]. It was evident that in this study, as the pH of the tea rhizosphere soil increased, the CEC of the soil increased, which promoted the decomposition of the soil organic matter, and then increased the contents of AN, AP and AK in the soil and enhanced the soil's fertility.

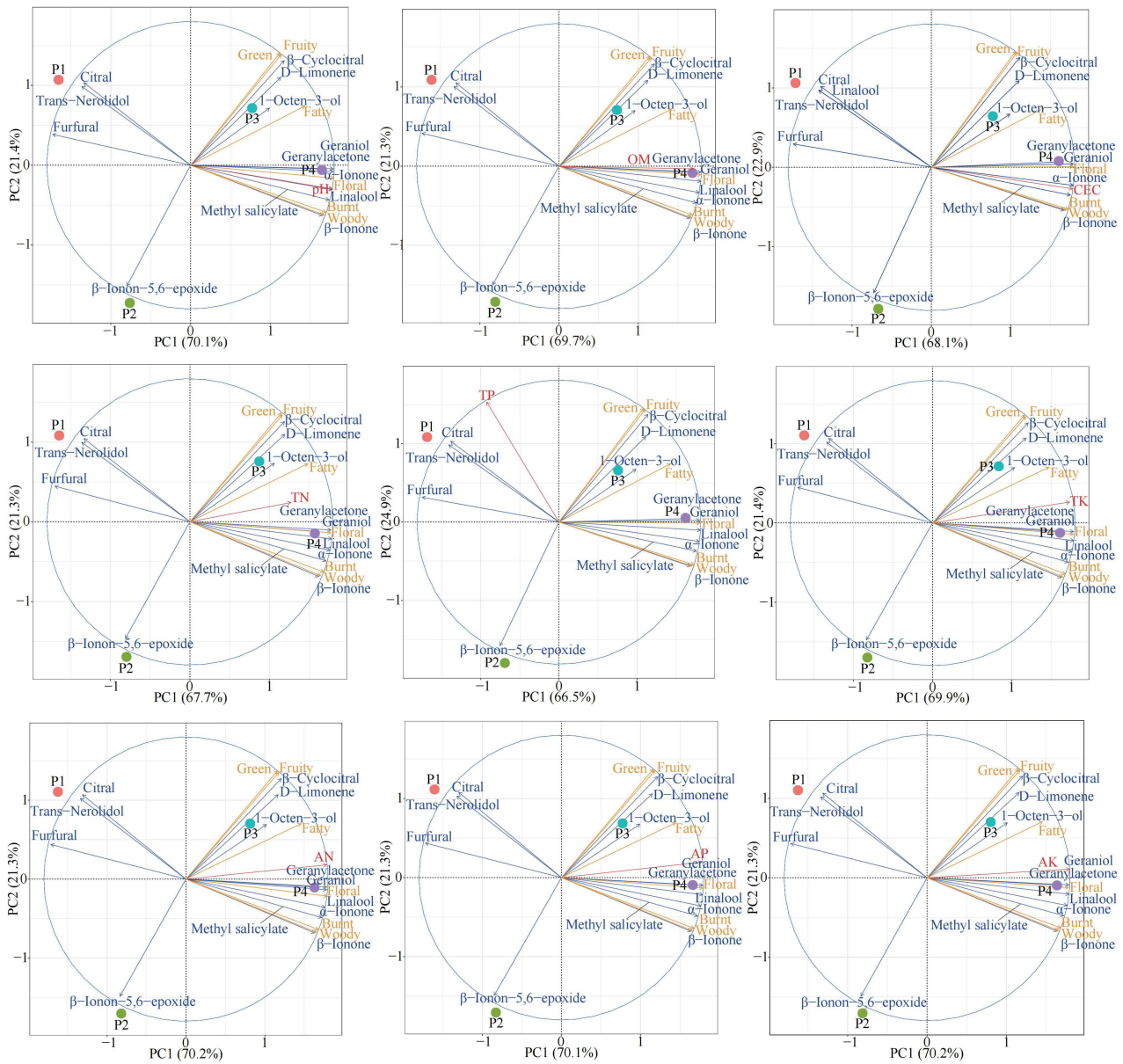


Figure 3. Principal component analysis of soil chemical indexes, key compounds and the odor characteristics of different samples. Note: P1—soil pH of 3.29; P2—soil pH of 4.13; P3—soil pH of 4.74; P4—soil pH of 5.32. pH—soil pH. OM—organic matter content; CEC—cation exchange capacity; TN—total nitrogen; TP—total phosphorus; TK—total potassium; AN—available nitrogen; AP—available phosphorus; AK—available potassium.

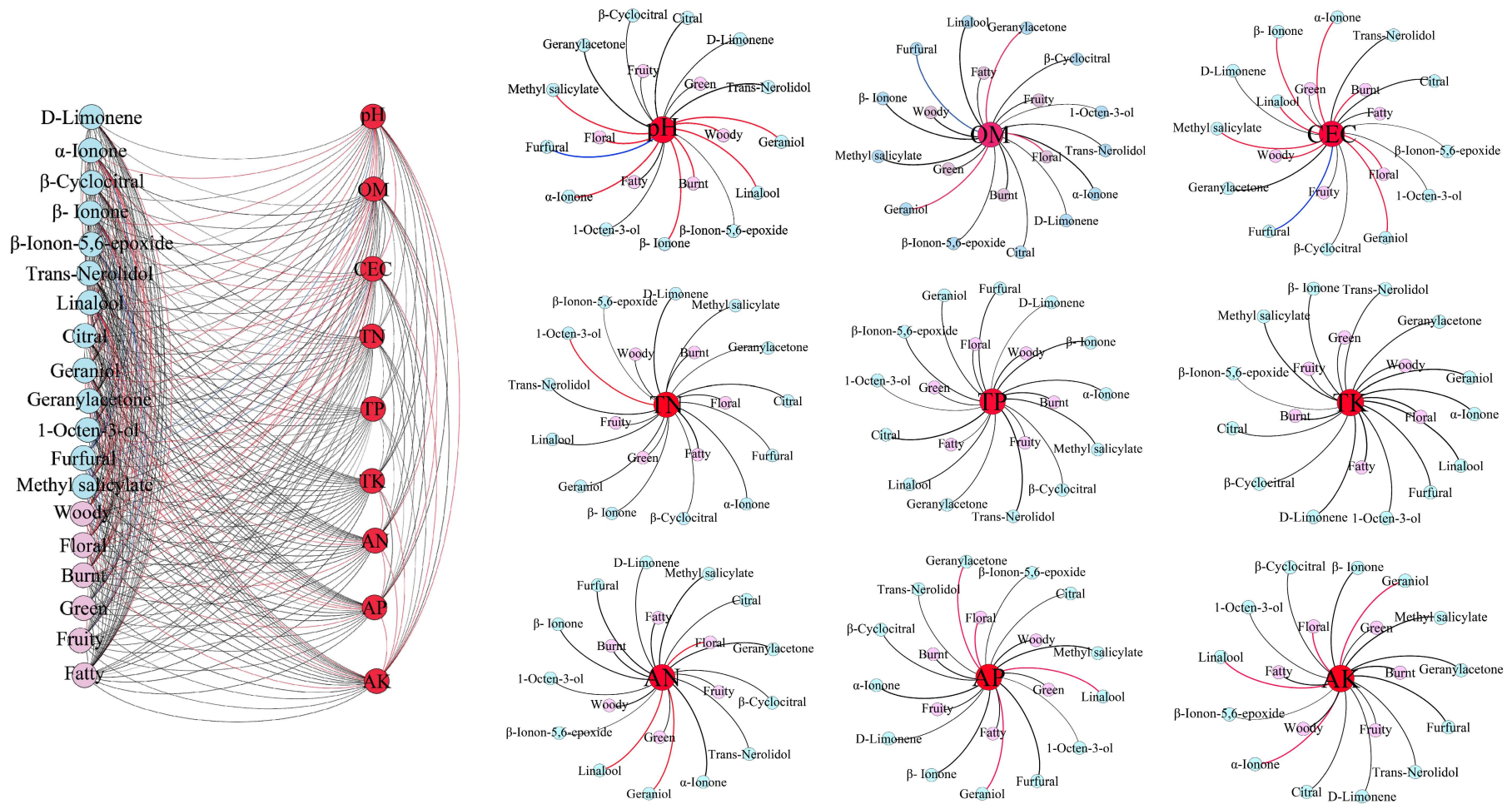


Figure 4. Interaction network of soil chemical indexes, key compounds and their odor characteristics based on correlation analysis. Note: — Significant positive correlation; - Significant negative correlation; — the correlation is not significant. — pH: soil pH; OM: organic matter content; CEC: cation exchange capacity; TN: total nitrogen; TP: total phosphorus; TK: total potassium; AN: available nitrogen; AP: available phosphorus; AK: available potassium.

A further analysis of the interaction of the soil chemical indexes, key aroma compounds and odor characteristics showed (Figure 4) that the soil pH and CEC were positively correlated with geraniol, α -ionone, β -ionone, linalool, methyl salicylate, floral, wood characteristics and burnt characteristics; the soil OM was positively correlated with geraniol, geranylacetone and floral characteristics; there was a significant positive correlation between the soil TN and 1-Octen-3-ol; the AN content was significantly and positively correlated with geraniol, linalool and floral characteristics; the AP content was positively correlated with geraniol, geranylacetone, linalool and floral characteristics; and there was a significant positive correlation between the soil AK content and geraniol, α -ionone, linalool and floral characteristics; however, there was no significant correlation between the soil TP and TK contents, the key compounds and the odor characteristics. A further analysis found that the soil chemical indexes except for TP and TK were positively correlated with geraniol and floral characteristics. It was reported that when the soil pH was 4.5~5.5, it was suitable for planting tea trees, and when the soil pH was 5.0~5.5, it was the most suitable for tea tree planting [1,2]. In this study, it was found that P3 and P4 were beneficial to the formation of six odor characteristics of tea leaves, where P3 was dominated by fruity, green and fatty characteristics and their main contributing compounds were β -cyclocitral, D-limonene and 1-octen-3-ol, while P4 was dominated by floral, woody and burnt characteristics, with the main contributing compounds being geranylacetone, geraniol, linalool, α -ionone, methyl salicylate and β -ionone. Secondly, it was found that except for TP, the soil chemical indexes had the greatest influence on the odor characteristics and key compounds of the P4 samples. It was evident that with the increase in the soil pH, the soil chemical indexes changed significantly, which was conducive to the synthesis of key compounds in the tea leaves with odor characteristics such as floral, woody and burnt. It can be seen that after the pH of the tea rhizosphere soil changed, the chemical indexes (CEC, OM, AN, AP and AK) in the soil changed immediately, and the chemical indexes affected the synthesis of compounds and the formation of odor characteristics in the tea leaves. This effect is manifested by the fact that an increase in soil pH favors the synthesis of compounds with floral characteristics, especially geraniol, in tea leaves, which is conducive to the enhancement of the floral characteristics of tea.

4. Conclusions

This study analyzed the effect of soil pH on the chemical indexes of rhizosphere soil, soil fertility and the aroma quality of tea trees. The results showed that increasing the soil pH is beneficial to the accumulation of CEC, OM and available nutrients in the rhizosphere soils of tea trees, thus improving the fertility of the soil. The aroma of Tieguanyin tea is dominated by a floral flavor, and the formation of floral characteristics is mainly derived from geraniol. The content of geraniol in the tea leaves was significantly and positively correlated with the soil's pH, CEC, OM content and available nutrient content. It can be seen that increasing the soil pH is conducive to improving the fertility of tea tree rhizosphere soil, which is conducive to increasing the content of geraniol in the tea leaves and thus enhancing the floral characteristics of the tea leaves. The tea tree is an acidophilic plant; however, the lower the pH value, the better, and an appropriate pH value aids in the transformation of soil nutrients and improves soil fertility, thus improving the quality of the tea. Therefore, in the process of tea planting, pH changes in the tea plantation soil are important; a pH of 5 or so is more appropriate, and for acidified soil, organic fertilizers (plant organic fertilizers, mineral organic fertilizers or decomposed sheep manure organic fertilizers, etc.) can be used in conjunction with chemical fertilizers appropriately in order to increase the soil pH and thus improve the quality of the tea. Therefore, the optimization of soil pH in oolong tea plantations is useful for the production of high-quality tea and for improving the economic benefits of tea farmers.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/agriculture13091739/s1>, Table S1. Classification standard for soil

fertility. Table S2. Aroma threshold and odor characteristics of some aromatic compounds. Table S3. GC-MS analysis of volatile compounds from the leaves of tea tree planted in soils with different pH levels. Table S4. Odor activity value (OAV) analysis of key aroma compounds and the characteristic aroma of tea leaves when the soil pH is 3.29. Table S5. Odor activity value (OAV) analysis of key aroma compounds and the characteristic aroma of tea leaves when the soil pH is 4.13. Table S6. Odor activity value (OAV) analysis of key aroma compounds and the characteristic aroma of tea leaves when the soil pH is 4.74. Table S7. Odor activity value (OAV) analysis of key aroma compounds and the characteristic aroma of tea leaves when the soil pH is 5.32.

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