

Research Article

Quality Evaluation of Three Kinds of Hickories Based on Grey Relational Analysis and Entropy-Weight Theory

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In this paper, the nutritional ingredient, aroma component, and texture of three kinds of hickories, including American hickory, Chinese Linan hickory, and Chinese Hunan hickory, were tested by instruments. The quality of different hickory varieties was analyzed at three levels by using the grey entropy correlation analysis, namely, the single nutrient composition analysis; nutritional composition and texture analysis; nutrient composition, texture, and aroma analysis. Through the analysis of nutritional composition, American hickory gets the highest score (80.6945), followed by Linan hickory (74.9987), and Hunan hickory has the lowest score (58.5925). Through the analysis of nutrition composition and texture, Linan hickory has the highest score (80.89), American hickory is the second (71.77), and Hunan hickory is last (61.62). Through the analysis of nutrition composition, texture and aroma, Linan hickory has the highest score (75.91), followed by American hickory (74.17), and Hunan hickory has the lowest score (64.20). Finally, the comprehensive evaluation of Linan hickory quality index score is the highest. The main factors contributing to the high score of Linan hickory include superior fatty acid spectrum, aminogram and higher initial chewing hardness, moderate crispness of secondary chewing, optimal palatability, and unique aroma components ((S)-2-methyl-1-butanol, 3-methyl-2-pentene, (+/-)-2-methylbutyric acid methyl ester ethyl butyrate, ethyl 2-methylbutyrate, methyl phthalate, decene, (1S)-(-)- β -pinene). The research results provide a basis for consumers to understand the quality differences of different hickories.

1. Introduction

Hickory is a wild nut, a natural pollution-free green food, also one of the many varieties with high nutritional value of dry fruit. Hayes et al., using a series of data sets and statistical methods, studied unique fatty acids and polyphenols of walnut. The results show that walnut may be considered to be a safe potential nutrient or drug. For cardiovascular disease, age-related nervous system diseases and even cancer, people can often eat walnuts as part of a healthy diet [1]. At present, there are many kinds of hickories in the Chinese market, including hickories from America, hickories from Linan, and hickories from Hunan. All three hickories have a large market share, but people lack sufficient knowledge of their quality characteristics and differences. Lillywhite et al. investigated 1009 American consumers based on the Internet group survey, and examined the population of hickory

consumers, their nut nutrition knowledge and purchasing preferences. Most of the respondents could correctly identify various nuts while they could not determine the specific nutritional characteristics [2].

Consequently, our purpose is to find a suitable method to comprehensively evaluate their quality differences according to the ingredients of different kinds of hickories. Thus, consumers can understand the differences among various hickories and purchase them in terms of their preferences.

At present, the research on the quality of hickory mainly focuses on testing nutrient ingredient of hickory by chemical methods and simple evaluation and comparison of their quality. Esteki et al. used pattern recognition to classify and identify Iranian walnuts from different geographic locations by analysis of fatty acid fingerprint based on gas chromatography [3]. Li et al. used cable-gas chromatography to

measure the total fat content and fatty acid composition of thirty-seven kinds of walnut [4]. Zhai et al. compared and analyzed the content of mineral elements and essential amino acids in *Juglans sigillata* and *J. regia* walnut kernel, examined their influence on human health, and sorted the mineral and amino acid contents [5]. Yi et al. established an infrared spectroscopy prediction model and measured and evaluated the moisture, protein, and fat content of walnut powder [6]. Prado et al. checked for the chemical composition of fatty acid, tocopherol, total oxidation stability index of phytosterol and peroxide value, analyzed composition, color and luminosity of hickory shell, tested extracts of total phenol, condensed tannins and antioxidant activity for hickory nut [7]. In addition to testing hickory kernels, some scholars have conducted research on hickory derivatives. Medina-Juarez et al. evaluated the phenolic content, total flavonoid content, concentrated tannin content, and antioxidant capacity of two varieties of hickories extract oil [8].

Physical properties of walnut kernels also have an effect on consumers' selection. Gharibzahedi et al. studied the differences in chemical, physical, and mechanical properties of three varieties Persian walnuts (Toyserkan, Chaboksar, and Karaj), which is mainly due to the individual characteristics of these varieties as well as the environment and cultivation conditions. The data obtained from these differences can be used for harvesting, transportation, sorting, and packaging [9].

Objective examination and comparison is difficult to reflect consumers' subjective feelings. There are flavor differences among different varieties of hickories. Magnuson et al. discussed the sensory differences in raw and baked eight kinds of hickory [10]. Miller and Chambers also evaluated seven black walnut varieties by sensory analysis. The trained seven members in a group developed a set of vocabulary for the black walnut and rated the sample of 22 flavor attributes [11].

Although many scholars have studied the nutrient ingredients of hickories, there is little deep and comprehensive research on the quality of hickories. The comprehensive evaluation methods, such as grey correlation degree (GCD), coefficient of variation method (CVM), analytic hierarchy process (AHP), fuzzy comprehensive evaluation (FCE), and DEA, have been applied in other fields, but there are few studies in the field of nuts or crops. For example, Veisi et al. used AHP to establish an ethics-based approach for sustainable agricultural indicator evaluation [12]. Abdollahzadeh et al. applied AHP to select management strategies of rice stem borer [13]. Yang and Mak proposed a multilayer FCE method which provides a classroom acoustic environment evaluation model to make reasonable sound processing suggestions for colleges and universities and improve the sound quality of the educational environment [14]. Chen et al. made comprehensive evaluation of environmental and economic benefits of anaerobic digestion technology for integrated food waste biogas plants based on fuzzy mathematical model [15]. Li et al. analyzed China's agricultural total factor energy efficiency based on the DEA and Malmquist indices [16]. Kao et al., based on dynamic and network DEA model, evaluated the cloud service industry.

For the cloud service industry, three NDEA models were built and solved by using multiobjective programming techniques [17]. Sun combined grey relational analysis and entropy models to empirically evaluate business performance [18].

As is discussed above, scholars have conducted many research studies about nutrients, volatile substances, and physical structure on hickory. However, few were paid attention to comprehensive evaluation. Consequently, our contribution is that the quality of hickory, nutrition ingredients, texture, and aroma are evaluated comprehensively by grey correlation analysis and entropy, which will provide buying reference for consumers.

The structure of this paper is as follows: in section 2, the experimental materials and research methods are presented. In section 3, the nutritional components, texture, and aroma of three kinds of hickories were analyzed, and the important indexes affecting the quality difference of hickories are discussed. The last part draws the conclusion of this paper.

2. Materials and Methods

2.1. Sample Collection. Samples are divided into Linan hickory, Hunan hickory, and U.S. hickory, from Qingliangfeng town of Linan, Huaihua of Hunan, and the United States of America. Each species is taken by 10 kg. On this basis, according to weights of samples, they are divided into three categories—big seeds, medium seeds, and small seeds, then their quantity and proportion are calculated, respectively. Concrete layering is shown in Table 1.

In order to calculate the appearance index of hickories, a total of 259 samples of hickories were drawn. Finally, according to the smallest sample size, 45 samples were taken for each variety. As experimental data, the distribution of samples is shown in Table 2.

2.2. Sample Detection Method. The quality of hickory was studied mainly from the aspects of nutrition, aroma, and texture. The corresponding index data were obtained by instrumental analysis. The quality indicator system of hickory is composed of three parts: nutritional composition, aroma, and texture. With a total of 81 indicators, among which 21 indicators were selected from nutritional composition, 50 indicators were selected from aroma, and 10 indicators were selected from texture.

For nutrients, according to GB 5009.124-2016, GB 5009.168-2016, GB 5009.3-2016, GB 5009.6-2016, GB/T 15686-2008, etc. (China Criterion), the fatty acids of Linan hickory, U.S. hickory, and Hunan hickory were tested by Zhejiang Gongzheng Testing Center Inc. (the third party inspection institution), a total of 14 kinds of fatty acid monomer components were detected. According to the detection for free amino acids and hydrolyzed amino acid of three kinds of original seeds of Linan hickory, American hickory, and Hunan hickory, 15 amino acid monomer components were detected. Excluding some indicators with less content, 21 nutrient indicators were selected.

TABLE 1: Laying data of samples.

Variety		Big seeds	Medium seeds	Small seeds	Sum
Linan hickory	Quantity	316	1630	730	2676
	Proportion	11.81%	60.91%	27.28%	
Hunan hickory	Quantity	153	458	769	1380
	Proportion	11.09%	33.19%	55.72%	
U.S. hickory	Quantity	147	1346	273	1766
	Proportion	8.32%	76.22%	15.46%	

Seed is classification by its weight. For Linan hickory, big seed >4.2 g, medium one (2.5, 4.2), and small one <2.5. For Hunan hickory, big one >10 g, medium one (7.14, 10), and small one <7.14. For American hickory, more than big one ≥ 6.5 g, medium one (4.3, 6.5), and small one <4.3.

TABLE 2: Source and distribution of samples.

Variety	Big seeds	Medium seeds	Small seeds
Linan hickory	5	28	12
U.S. hickory	4	34	7
Hunan hickory	5	15	25
Sum	14	77	44

For aroma, Linan hickory, American hickory, and Hunan hickory were fried by the same process. By SPME (solid-phase microextraction) and GC-MS (gas chromatography-mass spectrometry) analysis, 50 kinds of aromatic components were identified, including 3 kinds of aldehyde, 7 kinds of alcohol, 9 kinds of olefin, 6 kinds of ketones, 9 kinds of olefin, 6 of ester, 5 of aromatic hydrocarbon, 1 kind of steroids, 1 kind of furan, 1 kind of monoterpene, 1 kind of alkyne, and 1 kind of carboxylic acid.

For texture, five kinds of probes were selected for texture analysis, namely 1/2 shearing head, three-point bending special probe, P2E puncture probe, P100 pressure plate probe, HDPVB probe, and the simulated chewing scheme was used to test and extract data.

2.3. Research Method. This paper analyzed the grey correlation degree of hickory quality based on the entropy weighting method. We used grey correlation analysis to calculate the correlation degree of each hickory as the hickory quality score because the grey correlation analysis does not require too much sample size, nor does it require typical distribution rules. Also, the computation amount is less, whose results are consistent with qualitative analysis results. Considering that the subjective weight method will artificially affect the results of the index, we used the objective assigning method to determine the weight of each index in evaluating the quality of different hickories, that is, the entropy weight method. The calculation steps are as follows:

- (1) Establish the original evaluation matrix. According to the index system (nutrient, aroma, and texture), an $m \times n$ original evaluation matrix is established that m is the evaluation object and n is the evaluation index. X_{ij} represents the index value of the j th evaluation index of the i th evaluation object, and the original evaluation matrix is shown in Equation (1),

$$X = \begin{bmatrix} x_{11} & \cdots & x_{1n} \\ \vdots & x_{ij} & \vdots \\ x_{m1} & \cdots & x_{mn} \end{bmatrix}, \quad (1 \leq i \leq m, 1 \leq j \leq n). \quad (1)$$

- (2) Set up the reference sequence R_0 . Set the length of R_0 consistent with the number of columns in the evaluation matrix of $m \times n$. The reference sequence R_0 is the row vector composed of the ideal optimal values of each index. Add the reference sequence to row 0 of the original evaluation matrix to form a new evaluation matrix.

$$R_0 = \{x_{01}, x_{02}, \dots, x_{0j}\}. \quad (2)$$

In (2): x_{0j} is the optimal value of the j th column.

- (3) Calculate the correlation coefficient. As a reference sequence R_0 , calculate the correlation coefficient r_{ij} of each index of three kinds of hickories according to the following Equation (4):

$$r_{ij} = \frac{\min_i \min_j |x_{0j} - x_{ij}| + \mu \max_i \max_j |x_{0j} - x_{ij}|}{|x_{0j} - x_{ij}| + \mu \max_i \max_j |x_{0j} - x_{ij}|}, \quad (3)$$

where μ is the discrimination coefficient, $\mu \in [0, 1]$, take 0.5.

- (4) Calculate the weight. According to the theory of entropy weight method, the entropy value H_j of the j th index is calculated by Equation (4). Also, the weight of the j th index, ω_j is calculated by Equation (5),

$$H_j = -k \sum_{i=1}^m f_{ij} \ln f_{ij}, \quad 1 \leq j \leq n, \quad (4)$$

$$\omega_j = \frac{1 - H_j}{n - \sum_{j=1}^n H_j}, \quad 1 \leq j \leq n, \quad (5)$$

where in Equation (4), $k = (1/\ln n)$ and $f_{ij} = x_{ij}/\sum_{i=1}^m x_{ij}$, when $f_{ij} = 0$ and $f_{ij} \ln f_{ij} = 0$,

where in Equation (5), $0 \leq \omega \leq 1$ and $\sum_{j=1}^n \omega_j = 1$.

- (5) Calculate the comprehensive score. The score Y_i is calculated by Equation (6),

$$Y_i = \sum_{j=1}^n (r_{ij} \times \omega_j). \quad (6)$$

3. Results and Discussion

In the study of the hickory quality, everyone feels different, some people pay attention to nutrition; some people are attracted by the aroma; some people also pay attention to the taste. Therefore, our study judges the quality of hickories from three levels: firstly, considering nutrients and providing consumers with a reference on nutrients. Secondly, considering the nutrients and texture, namely taking into account the nutrients and stimulating the crisp chewing

perception of hickories. Finally, making a comprehensive analysis to the effects of nutrient, texture, and aroma on the quality of hickories as reference for consumers.

3.1. Consider Nutrients Only. The fatty acid and amino acid components of the nutrient components were detected, and a total of twenty-nine indicators were detected. Twenty-one kinds of indexes were selected after excluding the minimal components and other factors. According to the calculation of Equation (6), American hickory has the highest score, with the evaluation score of 80.6945, followed by Linan hickory with 74.9987, and Hunan hickory with the worst score is 58.5925 (see Table 3).

The sorting results in Table 3 are mainly due to the following reasons:

According to the detection for fatty acid ingredient of three raw seeds—Linan hickory, American hickory, and Hunan hickory—there are 14 kinds of fatty acid monomer components. The main ingredients include α -linolenic acid, linoleic acid, oleic acid, palmitic acid, and stearic acid. Also, the remaining components are minor components. The functional oil components detected are α -linolenic acid, linoleic acid, and oleic acid. Among the three kinds of hickories, the content of linoleic acid and oleic acid in American hickory is the highest, 14.10% and 38.20%, respectively. Its content of α -linolenic acid (0.98%) is slightly lower than that of Linan hickory and significantly higher than that of Hunan hickories. In addition, American hickory has the highest content of hickory palmitic acid (3.88%) and stearic acid content (1.44%). Above indicators reflect the good nutrient quality and health effects of U.S. hickory.

The free amino acid with the tannins and other taste substances together form the taste index of the hickories glycine, alanine, serine, and aspartic acid have obvious sweet taste, and the aspartic acid has a certain umami taste. Lysine, arginine, histidine, phenylalanine, tyrosine, leucine, isoleucine, and methionine have a certain bitter taste. Also, proline mixes a certain sweet and bitter tastes. The content of sweet amino acid in three kinds of hickories is 0.0503 mg/100 g (Linan hickory), 0.0376 mg/100 g (Hunan hickory), and 0.0375 mg/100 g (American hickory), respectively. The content of bitter amino acid is 0.0574 mg/100 g (Linan hickory), 0.0611 mg/100 g (Hunan hickory), 0.0465 mg/100 g (American hickory), respectively. Therefore, the sweet and bitter amino of American hickory are both the lowest.

The pericarp of the nut has a strong astringent taste, which comes from the tannin. Compared with Linan and Hunan hickories, American hickory has the least amount of tannin. After the same processing technology, it has better quality in taste than Linan and Hunan hickories. Among the 21 nutritional indicators, the highest proportion of weight was fat content, accounting for 0.063, followed by linoleic acid content, accounting for 0.061. The content of these two indicators of American hickory is far greater than that of Linan and Hunan hickories. The rich unsaturated fatty acids in nuts protect the cardiovascular system and help the body slow down aging. Figure 1 shows the visualized weights of 21 indicators and their descending order.

TABLE 3: Scores only considering nutrients.

Variety	Evaluation score
Hunan hickory	58.59247
Linan hickory	74.99874
U.S. hickory	80.6945

3.2. Consider Nutrient Composition and Texture. Considering nutrients and textures, there are thirty-one indicators. Compared with the analysis of only a single nutrient, ten texture indicators were added. The highest core was Linan hickory, with an evaluation score of 80.89, followed by American hickory with 71.77, and the last for Hunan hickory with 61.62, which indicates that Linan hickory has a crisper taste compared with American and Hunan hickory.

The sorting results in Table 4 are mainly due to the following reasons:

The weights of the three kinds of hickories considering nutrients and texture are shown in Figure 2. The higher weights are three-probe HDPVB hardness, three-probe P2 crispness, and fat. The special probe for three-point bending tests the brittleness of the samples by a three-point bending fracture. One indicator obtained by the three-point bending probe is hardness (brittleness). The hardness sorting of the test data of the three kinds of samples is as follows: Linan (20.52 N) > Hunan (19.12 N) > U.S. (16.04 N). It can be observed from the above data that the shearing stress to break Linan hickory is the highest, that of Hunan is middle and that of U.S. is the smallest, which reflects the highest hardness of Linan hickories, followed by Hunan and American hickories. The texture of American hickory is the softest. The test data of the probe indicates that Linan hickory has a large initial chewing hardness, and it is weaker than Hunan and American hickory in the brittleness of the first chewing.

3.3. Consider Nutrients, Texture, and Aroma Comprehensively. A total of eighty-one indicators were considered, including nutrition, texture, and aroma. Linan hickory scored 75.91, slightly higher than American hickory score of 74.17 and Hunan hickory score of 64.20. After adding aroma index, American hickory narrowed the gap with score of Linan hickory, which shows that American hickory has a unique aroma (see Table 5).

The sorting results in Table 5 are mainly as follows:

It is high for entropy weights of aroma component in hickories, and the total proportion accounts for 0.752397, which has an important influence on the evaluation of hickory quality. American hickory differs from Linan hickory and Hunan hickory in that it has a unique aroma different from others. However, Linan hickory and Hunan hickory also have their own unique aroma (see Figure 3).

According to the characteristic aroma that its value (aroma component content/threshold value) is greater than 1, it is determined that the unique characteristic aroma components for the American hickory are naphthalene, 4-methyl-3-pentenoic acid, tridecane, furfural, 2,6,6-

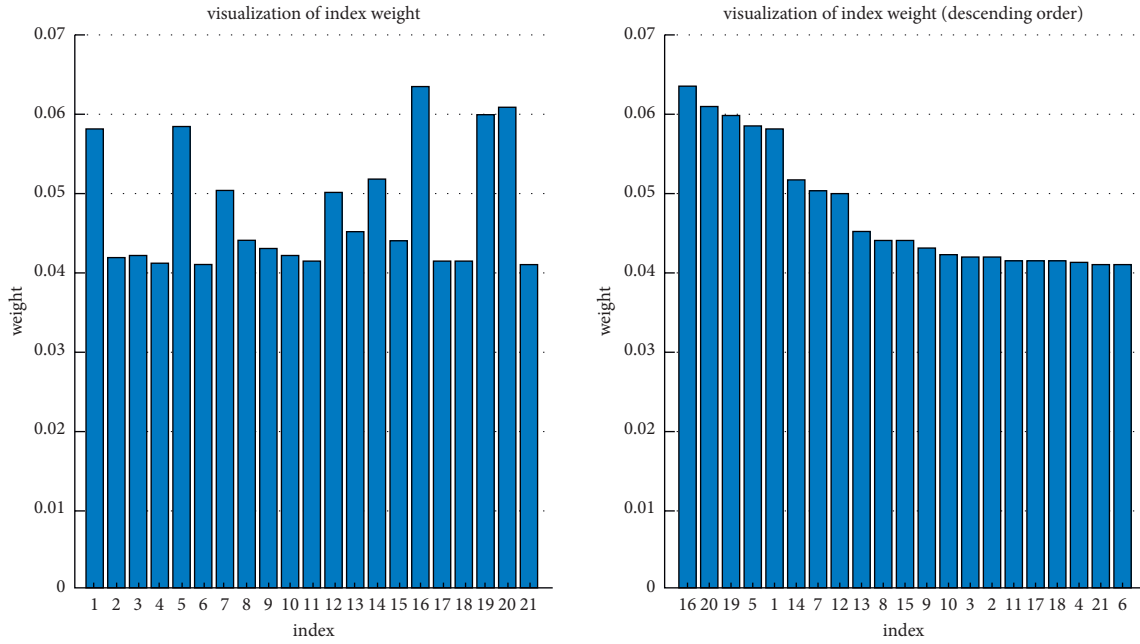


FIGURE 1: The weights of 21 indexes of three kinds of hickories with single nutrient component.

TABLE 4: Scores considering nutrient and texture.

Variety	Evaluation score
Hunan hickory	61.62219
Linan hickory	80.89055
U.S. hickory	71.77297

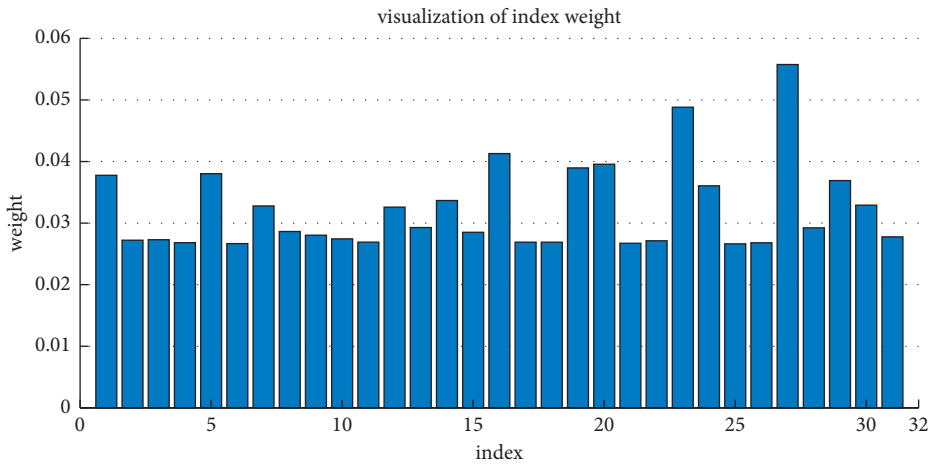


FIGURE 2: The weights of 31 indexes of three kinds of hickories with nutrient and texture.

TABLE 5: Scores of three kinds of hickories considering nutrition, texture, and aroma.

Variety	Evaluation score
Hunan hickory	64.20345
Linan hickory	75.90734
U.S. hickory	74.17492

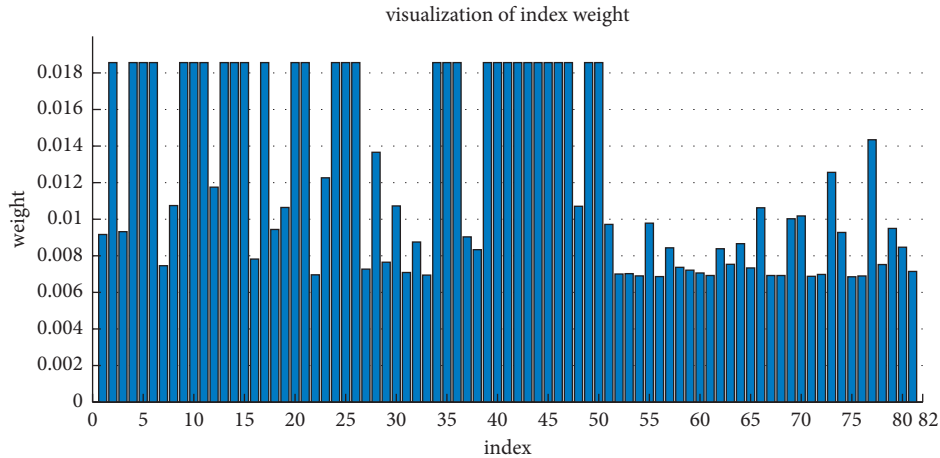


FIGURE 3: The weights of 81 indexes of three kinds of hickories with nutrition, texture, and aroma.

trimethyl-2-cyclohexene-1,4-dione, 2-propyl-1-heptanol, butyl butyrate, 4-methyldodecane, 2-methyl-1, 1'-biphenyl, tetradecane, 2,6-di-tert-butylphenylhydrazine, and diphenylmethane.

The unique aroma components of Linan hickory are (S)-2-methyl-1-butanol, 3-methyl-2-pentene, (+/-)-2-methylbutyric acid methyl ester ethyl butyrate, ethyl 2-methylbutyrate, methyl phthalate, decene, and (1S)-(-)- β -pinene.

The unique aroma components of Hunan hickory are 2-methyl-2-heptanol, 1,1'-((1-methylethylene)diethylene(oxygen))dibutane.

4. Conclusions

Our research helps consumers understand the quality of the hickories. The research report of quality is published in the media institutions by government annually. Consumers can make decisions of purchasing according to the quality scores. In this paper, we conducted a study from three aspects, and the main conclusions of the study are as follows,

- (1) Nutrient: compared with Linan hickories and Hunan hickories, the content of functional oils in American hickories is higher, and the fatty acid spectrum is superior. At the same time, the content of tannins with astringency is the lowest, and amino acid contents is also better.
- (2) Texture: evaluating through five kinds of texture probes from different angles, it is concluded that, compared with Hunan hickory and American hickory, Linan hickory has higher initial chewing hardness, moderate crispness of secondary chewing, and optimal palatable chewing. In terms of texture taste, Linan hickory is superior to American hickory and Hunan hickory.
- (3) Aroma: relative to the Linan hickory and Hunan hickory, unique aroma composition of American hickory is naphthalene, 4-methyl-3-pentene acid, tridecane, decanal, 2,2,6-trimethyl-2-cyclohexene-1,4-dione, 2-propyl-1-heptanol, n-butyl butyrate, 4-methyldodecane, 2-methyl-1,1'-biphenyl, tetradecyl,

2,6-di-tert-butyl-p-benzoquinone, and diphenyl methane. Because of unique aroma, American hickory is different from Linan hickory and Hunan hickory. However, it also provides a reference for consumers to choose their preferred taste.

- (4) The quality index system of hickory is composed of three parts, namely nutrition, texture, and aroma. The weight of the aroma is 0.752397, relatively large, and that of the nutrition is 0.165035, and that of the texture is 0.082568. The weight of the aroma is more than 75%, which determines the special quality of a kind of hickory to some extent. American hickory has outstanding aroma and superior nutrition. Linan hickory has a crisp taste and is also loved by consumers.

The experimental analysis in this paper is based on an objective evaluation method. It does not consider the subjective feelings from the perspective of consumers. On the other hand, only the grey correlation analysis method is applied. There are no other methods to do more comparisons with it. Also, it still needs studying whether other methods can be used to achieve the same conclusion or not. In the future, we will continue to improve our methods to solve more and more problems.

Data Availability

The data can be downloaded in <https://figshare.com>, DOI: 10.6084/m9.figshare.13084997.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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