Review Article
Food Science and Technology

ISSN 0101-2061 (Print)
ISSN 1678-457X (Online)

DOI: https://doi.org/10.1590/fst.08222

Research progress in the application of stable isotope and mineral element analysis in tracing the geographical origin of Chinese medicinal materials

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Abstract

The Geographical Origin of Chinese medicinal materials plays an important role in guaranteeing the authenticity of Chinese medicinal materials, ensuring the quality and efficacy of Chinese medicinal materials, maintaining the market order, and reducing the risk of medical accidents caused by counterfeit Chinese medicinal materials. In recent years, there have been more and more research papers on tracing Chinese medicinal materials by stable isotope and mineral element analysis combined with chemometrics. Using stable isotope and mineral element analysis to trace traditional Chinese medicinal materials has the advantages of high accuracy and strong reliability. Through chemometrics methods, medicinal materials from different origins can be classified, distinguished, and modeled, and finally the purpose of origin traceability can be achieved. Based on the introduction of the basic principles of stable isotopes and mineral elements, this article systematically analyzes the research and application of relevant stable isotope and mineral element analysis techniques in the origin traceability of Chinese medicinal materials in the past 10 years, and proposes the existing problems in origin traceability of Chinese medicinal materials. This article aims at providing reference for the origin traceability, confirmation and supervision of Chinese herbal medicines, and promoting the establishment and improvement of the traceability technical system of Chinese herbal medicines.

Keywords: Chinese medicinal materials; geographical origin; stable isotope; mineral element analysis.

Practical Application: To provide methodological and data analysis ideas for the application of stable isotopes and mineral elements in the traceability technology of Chinese medicinal materials.

1 Introduction

The origin of Chinese medicinal materials is closely related to the quality of traditional Chinese medicine. The quality, efficacy, and toxicity of the same medicinal materials come from different regions may be different; and the advantages and disadvantages of the quality directly affects the treatment of Chinese medicine. China has a vast territory, which is spanning multiple temperature zones from south to north with the complex and diverse terrain and environmental conditions. The doctors of all generations had summarized the relationship between drug changes and geographical environment, and proposed the theory of 'genuine medicine' based on the summarization. It refers to the best selected after long-term clinical application of traditional Chinese medicine and produced in specific regions; with higher quality, better curative effect and higher popularity than the same traditional Chinese medicine produced in other regions. Therefore, it is necessary to identify the same Chinese herbal medicine produced in different regions to ensure that the authenticity of traditional Chinese medicine (Liao et al., 2014; Wang et al., 2015; He et al., 2020; Xu et al., 2021a). Generally, studies on the identification of Chinese herbal medicines and origin are the analyzation and comparation on the appearance, ingredient content and genetic differences of Chinese herbal medicines for different geographic sources (Xiao et al., 2009). At present, most of the identification techniques for producing areas of Chinese medicinal materials in China are based on character identification, that is, to judge the authenticity of medicinal materials by describing the identification characteristics of the four major characters of medicinal materials: shape, color, smell and taste (Yuan et al., 2017). Character identification method mainly depends on the experience identification, namely, the identifiers usually judge the appearance and its internal quality of Chinese medicinal materials through the past experience. But this method is greatly affected by subjective factors, which requires the identifiers to have rich identification experience, and can only intuitively identify whether it is a qualified medicinal material similar to the main producing areas, and cannot accurately identify the traditional Chinese medicine from different producing areas (Chen et al., 2012). Advance in the rapid development of modern society, high-tech scientific and technological means have been continuously applied to the field of origin identification of Chinese medicinal materials, including high performance liquid chromatography (Sun et al., 2018), infrared spectroscopy (Yu et al., 2021), fluorescence spectroscopy (Hu et al., 2018), and even more advanced technologies, such as electronic nose technology (Liu et al., 2021; Wang et al., 2021a) and DNA fingerprint analysis technology (Wu et al., 2016). However, organic components in Chinese herbal medicines depends on various conditions (e.g., planting year, climatic

Received 02 Jan., 2022
Accepted 20 Feb., 2022
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conditions, geographic location, and soil composition), it is limitative to determine the origin of medicinal materials by the analysis of organic components, but the method of mineral elements and stable isotope analysis can better reflect the plant growth conditions (Drivelos & Georgiou, 2012). In addition, stable isotope and mineral elements analysis techniques have the advantages of fast analysis speed, high precision and remarkable effect in tracing the origin of Chinese medicinal materials. This has a promoting effect on maintaining the market order, enhancing the market competitiveness and enhancing patients' trust in the efficacy of Chinese medicinal materials (Wang et al., 2015). In order to promote the application of these two technologies in the field of traditional Chinese medicine, based on extensive domestic and foreign literature review, this paper reviewed and prospected the application of stable isotope and mineral element technology in the traceability and identification of traditional Chinese medicine.

2 Stable isotope technique

2.1 Principle of stable isotope technique

Isotopes refer to elements with the same proton number and different neutron numbers. A group of nuclides in the same position in the periodic table of elements are called isotopes. There are similarities and differences between them. Using the dissimilarity between isotopes, they can be separated and analyzed (Guo et al., 2006). Due to the continuous material circulation and energy exchange between organisms in nature and the external natural environment, the isotope in the body will produce natural fractionation effect under the influence of climate, water and soil factors, so that substances from different regions produce natural abundance differences, which can reflect the growth environment of organisms. Stable isotope ratio is a natural attribute of organisms, which is closely related to their growth environment and cannot be changed with the addition of chemical agents. Therefore, stable isotope information can be used to directly trace the origin of organisms (Guo et al., 2007). Stable isotope ratio analysis was first applied to measure the age determination in geological science. It has been applied to tracing the geographical origin of organic substances in biochemistry, archaeology, petroleum chemistry and geochemistry (Li et al., 2016). At present, there have been many studies at home and abroad using stable isotope analysis technology to identify the adulteration of ingredients or trace the origin, and have been successfully applied in fruit juice, tea and Chinese herbal medicine, etc (Lei et al., 2021). Isotope ratio mass spectrometry (IRMS), it is the most commonly measurement method of stable isotope ratio. There are two types of IRMS, that is, continuous stream IRMS and dual input IRMS, for dual input IRMS, gas is offline; and for continuous flow IRMs, gas is prepared online. Continuous stream IRMS can be coupled to another interface, such as element analyzer-isotope ratio mass spectrometry (EA-IRMS), gas chromatography -isotope ratio mass spectrometry(GC-IRMS) or liquid chromatography - isotope ratio mass spectrometry (LC-IRMS),of which, EA-IRMS is the most widely used in the traceability of Chinese medicinal materials (Pustjens et al., 2016). In the origin traceability, the most commonly used stable isotopes are C, H, O, N and S. The composition and changes of these isotopes are closely related to precipitation, altitude, geological environment, biological metabolism and cultivation methods (Liu et al., 2019). C stabilized isotopes include ^{12}C and ^{13}C, which are mainly related to the photosynthetic metabolism pathway of plants; H stable isotopes include ^{1}H and D, O isotopes including ^{16}O,^{17}O, and ^{18}O, H and O isotope ratios reflect the water source characteristics of plants; N stable isotopes including ^{15}N and ^{14}N, the $\delta^{15}N$ composition of plants is affected by various factors such as chemical fertilizers, climatic conditions, and soil conditions (Zhao et al., 2015; Danezis et al., 2016; Liu et al., 2019); the most common S isotopes are ^{32}S and ^{34}S, which are mainly affected by geological conditions, distance from the sea and some human factors; while, due to the different S sources of air pollution in different regions, the S isotopes composition in precipitation in different regions are significantly different. Some regions are easy to enrich ^{32}S, and some regions are easy to enrich ^{34}S (Tang et al., 2020).

2.2 Application of stable isotopes in origin traceability of Chinese medicinal materials

In recent years, stable isotope analysis technology has been widely used in the origin traceability of various Chinese herbal medicines, the plants mainly include rhizomes and fruits, and the animals are mainly sea cucumbers. Related research is shown in Table 1.

Rhizomes

Rhizome medicinal materials refer to underground rhizome medicinal materials with rhizome or a small amount of root or fleshy bulbs. Rhizome medicinal materials have high similarity in plant morphology, and it is difficult to identify their origin from appearance, but their origin can be identified by stable isotope ratios(Cen et al., 2021). Ding et al. (2014) firstly studied the effects of ecological factors on the $\delta^{13}C$ value values of different parts of Panax notoginseng, and it was found that latitude and annual precipitation were the dominant factors of $\delta^{13}C$ value variation of different parts, they provided a theoretical basis for the traceability of Panax notoginseng in different regions. He et al. (2015) also conducted similar research; from 49 ecological indicators, they screened out 8 major ecological factors that influence the $\delta^{13}C$ of main roots of Panax notoginseng, namely, the average temperature in July, the lowest temperature in January, latitude, longitude, annual average temperature, soil available zinc content, soil total phosphorus content and soil available potassium content. And analyzed the size, direction, decisive factors and restrictive factors of the eight ecological factors, they found that $\delta^{13}C$ of main roots of Panax notoginseng was mainly affected by temperature and longitude of origin. Li et al. (2021) researchs results showed that the fractionation effect of $\delta^{13}C$, $\delta^{15}N$, $\delta^{2}H$ and $\delta^{18}O$ values in different parts of Panax notoginseng was inconsistent. The δD value of each part of Panax notoginseng decreased with the increase of altitude and latitude, while, the $\delta^{18}O$ value was opposite. The interannual factors could be ignored in the traceability of Panax notoginseng origin. In addition to using a single isotope traceability, the joint traceability of multiple isotopes is also a common method. Liang et al. (2021) expanded the isotope analysis index, studied

Table 1. Origin Traceability of Chinese Medicinal Materials Based on Stable Isotopes.

samples	sources	key variables	statistical method	results	reference
Panax notoginseng	Guangxi, Yunnan,	$\delta^{13}C$, $\delta^{15}N$, $\delta^{2}H$, $\delta^{18}O$	ANOVA, SDA, Fisher discriminant analysis	overall discrimination rate about 84%;	Liang et al. (2021)
Ginseng	China, Korea,	δ^{13}, $\delta^{15}N$, $\delta^{2}H$,	F-test, boxplot	good distinction;	Horacek et al. (2010)
Dendrobium officinale	Guangdong, Guangxi, Guizhou, Yunnan, Zhejiang,	$\delta^{13}C$, $\delta^{15}N$, $\delta^{2}H$, $\delta^{18}O$, $^{87}Sr/^{86}Sr$	PCC, PCA, One-way ANOVA,	PCA can distinguish; Dendrobium officinale from different habitats;	Yang et al. (2018a)
Panax japonicus	Akita, Hubei, Yunnan, Sichuan, Guizhou,	$\delta^{13}C$, $\delta^{15}N$, $\delta^{2}H$, $\delta^{18}O$	SVM, LDA, BPN, One-way ANOVA,	accuracy of LD and BPN is 100%;	Zhang et al. (2020)
Amomum villosum	Guangdong, Guangxi, Yunnan,	$\delta^{13}C$, δD, $\delta^{15}N$	OPLS-DA, LDA	the accuracy of discriminant model is 89.7%	Zhong et al. (2021)
Lycium barbarum L.	Qinghai, Ningxia, Xinjiang,	$\delta^{13}C$, $\delta^{15}N$, $\delta^{2}H$, $\delta^{18}O$, $\delta^{34}S$	PCA, LDA, SVM, One-way ANOVA	the discrimination effect of SVM model is better than that of LDA model;	Lian et al. (2022)
Lycium barbarum L.	Gansu, Ningxia, Qinghai,	$\delta^{13}C$	ANOVA, LDA	LDA can distinguish *Lycium barbarum* L. from different provinces;	Meng et al. (2019)

Note: ANOVA = Analysis of variance; SDA = Stepwise discriminant analysis; PCC = Pearson's coefficient of correlation; PCA = Principal component analysis; One-way ANOVA = One-way analysis of variance; SVM = Support vector machine; LDA = Linear discriminant analysis; BPN = Back propagation neural network; LD = Linear discriminant; OPLS-DA = Orthogonal projection to latent structure-discriminant analysis; following is the same.

the values of $\delta^{13}C$, $\delta^{15}N$, $\delta^{2}H$ and $\delta^{18}O$ in the main roots of Panax notoginseng, and used Duncan multiple comparison analysis, Fisher discriminant analysis and stepwise discriminant analysis to process the data. The results showed that there were significant or extremely significant differences in the values of $\delta^{13}C$, $\delta^{15}N$, $\delta^{2}H$ and $\delta^{18}O$ between the producing areas. Based on the four stable isotope ratios, the average discrimination accuracy of the origin discrimination model was 84%.

Ginseng is a well-known Chinese herbal medicine in rhizomes. The application of stable isotope analysis in the traceability of ginseng origin has also achieved certain results. Micha et al. (2010) studied the $\delta^{13}C$, $\delta^{2}H$, and $\delta^{15}N$ values in Chinese and Korean ginsengs. The results showed that the $\delta^{2}H$ value in Chinese ginseng was significantly higher than Korean ginseng, and the difference of $\delta^{2}H$ value could be used to effectively distinguish Korean ginseng from Chinese ginseng. H isotope has a good effect on the origin traceability of ginseng in a large scale, and also has a good effect on the identification of ginseng in adjacent producing areas. Besides, for ginseng, in addition to the growth environment, the varieties may also affect its isotope composition. Chung et al. (2018) studied the values of $\delta^{13}C$, $\delta^{15}N$, $\delta^{18}O$ and $\delta^{34}S$ between different varieties in South Korea, the results showed that the interaction of geographical and varieties would significantly affect the ginseng $\delta^{13}C$, $\delta^{15}N$, $\delta^{18}O$ and $\delta^{34}S$ ($P < 0.001$). The difference between $\delta^{15}N$ and $\delta^{13}C$ has significant significance for ginseng to distinguish between different origins, in addition, Chung et al. (2017) also studied the influence of soil type and fertilizer type on $\delta^{13}C$, $\delta^{15}N$and $\delta^{34}S$ value of ginseng, found the $\delta^{15}N$ and $\delta^{34}S$ value were related to the type of soil and fertilizer.

Study on the origin traceability of other rhizome Chinese medicinal materials by stable isotope analysis, except panax notoginseng and ginseng; now, it is still in the stage of exploring effective traceability index. Yang et al. (2018a) determined the isotope ratios of Sr, C, H, O and N in Dendrobium officinale from different regions of China. The results showed that the Sr isotope ratio could be used to effectively distinguish Dendrobium officinale from Zhejiang and other producing areas. Zhang et al. (2020) studied the values of $\delta^{13}C$, $\delta^{15}N$, $\delta^{2}H$ and $\delta^{18}O$ in Panax japonicus from six producing areas, and used LDA, SVM and BPN to process the data. The results showed that $\delta^{13}C$ had obvious regional characteristics, which could be used to effectively distinguish the producing areas of Panax japonicus, and the accuracy of LDA and BPN was 100%.

Fruit

Fruit Chinese medicinal materials come from the whole fruit or part of the fruit, such as Amomum villosum, *Lycium barbarum* L. and other medicinal materials. Zhong et al. (2021) studied the values of $\delta^{13}C$, $\delta^{15}N$ and $\delta^{2}H$ in Amomum villosum from Guangdong, Guangxi and Yunnan, and used OPLS-DA to screen the values of $\delta^{13}C$ and δD with significant differences;they established a discriminant model for origin traceability by LDA, and the discriminant accuracy was only 76.9%, because of the selection of discriminant method, the element index of analysis and the geographical location of sample collection may affect the discriminant accuracy. Lian et al. (2022) studied the values of $\delta^{13}C$, $\delta^{15}N$, $\delta^{2}H$, $\delta^{18}O$ and $\delta^{34}S$ in 97 *Lycium barbarum* L. samples from 3 provinces in China; they used Box diagram, ANOVA and PCA to process the data, and used LDA and SVM to made a model. The results showed that LDA model and SVM model could effectively distinguish the origins, but the discriminant effect of LDA model was slightly worse than that of SVM. Besides the determination of overall isotope difference in samples,the determination of isotope distribution of specific compounds is also applied in the identification of origins. For example, Meng et al. (2019) analyzed the $\delta^{13}C$ values of volatile compounds (limonene, ligustrazine, saffron aldehyde, geranyl acetone and β-ionone) in the *Lycium barbarum* L. from Ningxia, Qinghai and Xinjiang. The results showed that the δ13C values of volatile compounds (ligustrazine, geranyl acetone and β-ionone) were the most important identification variables, and the accuracy of LDA model was 87.6%.

Animalia

In addition to plant-derived Chinese herbal medicines, animal-derived Chinese herbal medicines are also the characteristic components of traditional Chinese medicine, it has a wide range of applications in clinical medicine; and many studies have applied mineral elements and stable isotopes to the origin traceability of animal-derived Chinese herbal medicines. Zhang et al. (2017) studied the $\delta^{13}C$, $\delta^{15}N$ values and fatty acid profiles of sea cucumbers in northern China, and the results showed that there were significant differences in the $\delta^{13}C$, $\delta^{15}N$ values and fatty acid profiles of sea cucumbers in different regions. Sea cucumbers in different regions could be accurately distinguished by PCA and DA. It is proved that the combined analysis of stable isotope ratio and fatty acid map could be used as an effective means for origin traceability of marine products. Kang et al. (2019) studied the sea cucumbers from five different regions in China. By measuring the C, H, O, N contents and stable isotope ratios in sea cucumbers, it was found that the C, H, O, N contents and stable isotope ratios in sea cucumbers from different producing areas were significantly different ($P < 0.05$). The overall discrimination rate of PCA was 93.4%, which was better than that of LDA (89.5%). Li et al. (2014) analyzed the $\delta^{13}C$ and $\delta^{15}N$ values of Cordyceps sinensis in Qinghai, Yunnan, Sichuan and Nepal. The results showed that there was no discernible difference in the δ13C values of Cordyceps sinensis from different habitats, but the $\delta^{15}N$ value was fluctuated, the $\delta^{15}N$ value could be used to distinguish the habitats of Cordyceps sinensis generally.

3 Application of mineral element analysis in origin traceability of Chinese medicinal materials

3.1 Technical principle of mineral element analysis

As one of the basic components in organisms, the mineral elements are important substances involved in biological metabolism, but the organism itself cannot synthesize mineral elements; so, for plants, it must absorb the maintain various from soil, water, atmosphere or other else. The mineral element composition in plants is closely related to the element composition of their growth environment. The composition and content of mineral elements in different regions are different, resulting in the characteristic differences in the composition and content of mineral elements in plants from different regions. This characteristic difference could be used to study the origin traceability of Chinese medicinal materials. In general, in the practice of origin traceability, inductively coupled plasma optical emission spectrometer (ICP-OES) or inductively coupled plasma mass spectrometry (ICP-MS) are usually used to simultaneously determine a variety of constant, trace and trace elements, and elements with significant differences are selected as identification factors, and use artificial neural network, PCA and other chemometrics to distinguish production areas (Liu et al., 2019; Drivelos & Georgiou, 2012; Pranoto et al., 2021).

3.2 Application of mineral elements analysis technology in origin traceability of Chinese medicinal materials

In recent years, mineral elements analysis technology has been used in the studies of origin traceability of Chinese medicinal materials such as *Lycium barbarum* L., Panax notoginseng, and Medulla Tetrapanacis etc., we have achieved satisfactory results. Related research summary is shown in Table 2.

Table 2. Origin traceability of Chinese medicinal materials based on mineral elements.

samples	sources	key variables	statistical method	results	reference
Pueraria thomsonii	Shaanxi, Hubei, Guangxi	Mg, P, Co, Rb, Sr	PCA, SLDA, HCA	the overall accuracy rates of back substitution test and cross test are 97.7% and 93.0%, respectively;	He et al. (2021)
Medulla Tetrapanacis	Guangxi, Sichuan, Guizhou, Yunnan	Sn, Be, Na, As, Mg, Al, K, Ca, Mn, Fe, Co, Cu, Zn, Se, Si, Sb, Ba, Ti, Pb, Zr, Sr	PCA, DA, SLDA, K-NN	the correct resolution of KNN model training set and validation set are 100% and 93.33%;	Li et al. (2014)
Salvia miltiorrhiza	Shandong, Shanxi, Henan, Anhui	Na, K, Ca, V, Fe, Ni, Cu, As, Se, Mo, Cd, Pb, Th, U, Sc, Y, La, Eu	ANOVA, LDA, PCA	the accuracy of LDA regression and the success rate of cross validation are 100% and 85.5%;	Zhao et al. (2017)
Lycium barbarum L.	Ningxia, Gansu, Xinjiang	Mg, Fe, Ca, Zn, Cu, Mn, Al, Sr, Rb, Mo, Tb, Cd, Co, V, As, Cs, Pr, Dy, Gd	ANOVA, PCA, LDA	the accuracy rates of original group and cross validation group are 95.7% and 85.7%;	Zhang et al. (2016)
Lycium barbarum L.	5 regions of Zhongning, Ningxia	Cd, Ce, Co, Cu, Gd, Hg, Mg, Se, Zn, P	ANOVA, PCA, PLS-DA, Fisher	OPLS-DA is better than Fisher;	Kai et al. (2021)
Panax notoginseng	5 regions of Yunnan	Cu, Fe, Mn, Na, Cd, Pb, As	FA, PCA, CA, CDA	CA discrimination accuracy is 90%;	Zhang et al. (2021)

Note: PLS-DA = Partial least square discriminant analysis; FA = Factor analysis; CA = Cluster analysis; CDA = Canonical discriminant analysis; DA = Discriminant analysis; SLDA = Stepwise linear discriminant analysis; K-NN = k-Nearest Neighbor; HCA = Hierarchical cluster analysis; following is the same.

Rhizomes

Mineral elements in plants are related to soil environment, and the differences in soil between different regions will lead to differences in mineral elements in samples from different regions (Xiang et al., 2015). He et al. (2021) studied 22 mineral elements in Pueraria thomsonii and its planting soils in different regions; after the discriminant analysis, they screened 5 effective indicators for provenance traceability of Mg, P, Co, Rb and Sr and established a discriminant model. The total discriminant rates of back substitution test and cross test were 97.7% and 93.0%, respectively. In addition, it was found that Rb, Sr and Ti in soil were significantly positively correlated with the corresponding elements in Pueraria thomsonii ($P<0.01$), which proved that the feasibility of mineral element fingerprint combined with chemometrics in the origin traceability of Pueraria thomsonii.

The same chemometrics method has great differences in the origin traceability of different Chinese medicinal materials. CA as an example, Xu et al. (2021) studied the inorganic elements in Rhizoma cyperi from different producing areas in China, the results showed that Rhizoma cyperi could be clustered by CA according to the producing areas of East, Central, North, Southwest and South China, but it could not be accurately clustered by provinces. Zhou et al. (2020) also used the same analysis method to discriminate dry ginger from Yunnan, Guizhou, Sichuan, Shandong in China. The results showed that CA only clustered Shandong dried ginger into one group, Guizhou, Sichuan and Yunnan dried ginger into one group. CA was also used to identify the origin of Chinese medicinal materials, while other research results showed that CA had good origin identification ability. Jiang et al. (2020) analyzed 27 mineral elements in Radix Peucedani from different habitats in China (Anhui, Zhejiang, Guizhou, Chongqing). The results showed that the contents of Si, Ca and K in Radix Peucedani from different producing areas were significantly different, and CA could distinguish Radix Peucedani according to different provinces. Zhang et al. (2021) studied 11 elements in Panax notoginseng samples from 5 regions of Yunnan, and screened out 7 elements with significant differences among different producing areas by ANOVA. And then, they used PCA, CA and CDA to classify the main roots of Panax notoginseng, the accuracy of CA was as high as 90%. In summary, when the same chemometrics method is applied to the origin traceability of different Chinese medicinal materials, the discriminant effect is different, this may be related to the analysis elements and sampling points.

Chemometric methods are very important for origin traceability. For the same data, different chemometric methods have different results in origin traceability. Li et al. (2015) used PCA, CA, SLDA and KNN to classify the origins of Medulla Tetrapanacis. The content of mineral elements in the samples from different origins was KNN > SLDA > PCA > CA. Zhao et al. (2017) studied the content of 35 elements in Salvia miltiorrhiza from different producing areas, and used PCA and LDA to analyze the data. The results showed that PCA could only roughly distinguish the origin of the sample, but LDA could accurately distinguish all the samples. Therefore, there is no absolute advantages and disadvantages of different classification and pattern recognition methods in different situations, only evaluated it according to the actual distinction effect of the sample.

Fruit

Lycium barbarum L. is a traditional Chinese medicinal material, which healthy nutritional value has been widely recognized, and related products are sold at home and abroad. Zhang et al. (2016) analyzed the 20 mineral elements in 40 *Lycium barbarum* L. from different regions and different periods in ZhongNing, Ningxia. Combined with the traceable indicators screened by ANOVA and PCA, the results showed that the content of mineral elements in *Lycium barbarum* L. at different periods was basically stable; but there were visible interlocal diversities in 19 mineral elements. Based on these 19 variables, a linear discriminant model was established; the correct resolution of original group was 95.7%. and the correct rate of cross-validation group was 85.7%, preliminarily confirmed the feasibility of mineral elements for the origin traceability of *Lycium barbarum* L. in ZhongNing. Kai et al. (2021) also carried out the similar studies, and expanded the number of samples, determination elements and analysis methods; they determined 43 mineral elements in 111 samples from 5 production areas of ZhongNing, and then, established a discriminant model of production area based on Fisher discriminant analysis method. Based on OPLS-DA discriminant analysis method, the overall correct discriminant rates of two discriminant models were 82.0% and 91.89%, respectively. In addition, Kai et al. (2020) also studied the differences of mineral elements in varieties of *Lycium barbarum* L. in Ningxia. The results were similar to those before, the mineral elements of *Lycium barbarum* L. in different regions have different characteristics, which further confirmed that the mineral element analysis method was feasible for the origin identification of *Lycium barbarum* L.; on the other hand, there was no significant difference in the elements content of different *Lycium barbarum* L.; therefore, the origin traceability of *Lycium barbarum* L. was almost unaffected by the variety difference.

Animalia

Animals themselves cannot synthesize mineral elements and must be absorbed from the outside world. They mainly come from food and drinking water, and a small amount comes from the natural environment, such as air and soil. Therefore, animals in different regions have their own characteristics of mineral elements (2021). Studies have found that there are differences in the contents of individual mineral element in the sika deer's initial antler, two-branched antler, three-branched antler and regenerated antler. Among them, the contents of Zn and Mg in antler in Jilin were significantly lower than those in Shandong, and the contents of P and Ca in antler were also significantly different between them, these also confirmed the feasibility of using certain or certain elements for antler traceability (Wang et al., 2021b; Zhao et al., 2019). Wu et al. (2014) determined 14 mineral elements in the mactra chinenisis from 5 sampling points in Rushan Bay by ICP-MS. They established a four-factor model by CA and PCA, and its accuracy rate of the sample was 80%. Shao et al. (2016) also used the same analytical method to determine the mactra chinenisis from Jiao Zhou Bay in the subsequent studies; compared with the mactra chinenisis in Ru Shan Bay, it was found that the Hg content of heavy metal in Jiao Zhou Bay was higher than that in Ru Shan Bay, and the Al and AS values could be used as effective indicators for identification, A discriminant model with a correct discriminant rate of 93% was established by Al and As values.

4 Combined analysis of stable isotopes and mineral elements

At present, a lot of research results have been achieved by using stable isotopes or mineral elements alone to trace the origin of Chinese herbal medicines. However, each traceability technology has its own advantages and limitations. There is almost no analytical technology that can independently complete the origin traceability. Especially, when the accuracy of origin traceability technology is to be improved, the limitations of a single technology will be more prominent. In recent years, multi-technical and multi-parameter joint analysis method has gradually become a hotspot for tracing research in traditional Chinese medicine production, Joint analysis method can refine regional information, screen more characteristic analysis parameters, improve the accuracy of discriminant model, and achieve accurate and reliable origin traceability. Among them, the combined analysis of stable isotopes and mineral elements is a potential provenance traceability technology, which has achieved good results in the traceability of plant-derived products and animal-derived products (Sun et al., 2018). Table 3 summarizes the studies that the application of combined analysis of stable isotopes and mineral elements in traceability of Chinese medicinal materials in recent years.

4.1 Rhizomes

It is feasible to use the stable isotope value or mineral element content alone as the discriminant model variable in the traceability of Chinese medicinal materials, but the stable isotope combined with mineral element analysis method can improve the accuracy and stability of the origin identification. Du et al. (2020) studied the mineral elements and stable isotopes in coptis chinensis from different regions in China, and established the origin discrimination model of coptis chinensis based on Al, Sn, Mg, $\delta^{13}C$ and $\delta^{15}N$ as variables through the linear discriminant analysis and OPLS-DA. The correct discrimination rate of the predicted samples was 100%. Angelica sinensis is a perennial herb in China, the root system development and yield of Angelica sinensis produced in different regions were different, as were the content of trace elements and the medicinal effect (Yan et al., 2021). Yang et al. (2020) analyzed the $\delta^{13}C$, $\delta^{15}N$ values and 7 trace elements contents in angelica sinensis from Gansu, Sichuan and Yunnan. The results showed that the stable isotope values and mineral elements in angelica sinensis from different producing areas had their own regional characteristics. Using $\delta^{13}C$, $\delta^{15}N$ value and content as discriminant factors, the accuracy of FLDA back substitution and cross validation was 90% and 83%, and the accuracy of external samples was 63%. After adding seven trace elements, the accuracy of FLDA back substitution and cross validation increased to 100% and 91%, and the accuracy of external sample prediction increased to 100%. Yang et al. (2018b) also found that using stable isotope ratio can effectively distinguish Polygonum multiflorum origins but combined with element analysis can further improve the accuracy of provenance traceability model test. Mineral element analysis is better than stable isotope analysis in some Chinese

Table 3. Research on the origin traceability of Chinese medicinal materials based on stable isotopes and mineral elements.

samples	sources	key variables	statistical method	results	reference
Coptis chinensis franch	Sichuan, Shaanxi, Yunnan, Hubei, Chongqing	$\delta^{15}N$, $\delta^{13}C$, Al, Sn, Mg	PCA, LDA, OPLS-DA	LDA and OPLS-DA classification accuracy are both 100%	Du et al. (2020)
Angelica sinensis	Gansu, Sichuan, Yunnan	$\delta^{13}C$, $\delta^{15}N$, Mg, Mo,Cr, Mn, Fe, Ni, Zn	One-way ANOVA, FLDA	the accuracy of FLDA is 100%, the accuracy of cross validation is 91%, and the accuracy of external sample prediction is 100%;	Yang et al. (2020)
Polygonum multiflorum thumb	Anhui, Hubei, Sichuan	$\delta^{13}C$, $\delta^{15}D$, $\delta^{18}O$,Sc, V, Zn, Rb, Cd	One-way ANOVA, PCA, OPLS-DA, FLDA, CHAID	the accuracy of back substitution test, cross validation and external sample test for decision tree CHAID modeling analysis reaches 100%.	Yang et al. (2018b)
Atractylodes macrocephala	Anhui, Sichuan, Chongqing, Hunan,	$\delta^{18}O$, $\delta^{34}S$, Li, Dy, Sm, Er	one-way ANOVA, PCA, LDA	classification accuracy is 100%	Fu et al. (2020)
Polygonatum sibiricum	Anhui,Shanxi, Hebei,Guizhou, Sichuan,Yunnan	$\delta^{13}C$, $\delta^{15}N$, $\delta^{2}H$, $\delta^{18}O$, Sr, Be, U	one-way ANOVA,PCA,OPLS-DA	the accuracy of comprehensive discrimination is 100%;	Fu et al. (2021)
Lonicera japonica	Henan, Shandong, Hebei, Chongqing	$\delta^{13}C$, $\delta^{15}N$, $\delta^{18}O$, Cd, Cr, As, Hg, Se, Co, Ni, Sr, Fe	ANOVA, PCA, LDA	LDA cross validation accuracy is 95.7%;	Fan et al. (2018)
Semen Cassiae	Zhejiang,Hebei, Shandong,Shaanxi, Sichuan, Anhui, Hunan, Guangxi, Hainan	$\delta^{13}C$, $\delta^{15}N$, Ca, Mn, Y, Nd, Gd, Pr, La, Al, V, Li, As, Ba	one-way ANOVA, PCA, ANN	the overall accuracy of ANN model for the analysis of origin discrimination is higher than 90%;	Wang et al. (2020)

Note: FLDA = Fisher Linear Discriminant Analysis; ANN = artificial neural network.

medicinal materials. Fu et al. (2020) found that the accuracy of discriminant model based on different variables was different when using stable isotopes and mineral elements to discriminate the origin of atractylodes macrocephala in different habitats, the stable isotope binding mineral elements (100%) > mineral elements (98.67%) > stable isotope rate (88.17%).

Stable isotope and mineral element analysis method can identify the origin of Chinese medicinal materials, and also can identify natural growth and artificial planting of Chinese medicinal materials. Fu et al. (2021) analyzed the $\delta^{13}C$, $\delta^{15}N$, $\delta^{18}O$, $\delta^{2}H$, ^{87}Sr / ^{86}Sr and 48 multi-element characteristics of Polygonatum sibiricum from the main product areas. It was found that CA, ANOVA and OPLS-DA not only can identify from Polygonatum sibiricum different habitats, but also can effectively distinguish natural Polygonatum sibiricum from artificial Polygonatum sibiricum, and the accurate identification rate was 100%.

4.2 Flowers and fruit

Except for the rhizome Chinese medicinal materials, stable isotopes combined with mineral element analysis were used to determine the $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$ values and 18 mineral element contents in Lonicera japonica from different regions by Isotope Ratio Mass Spectrometry (IRMS), Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-OES) and Inductively Coupled Plasma Mass Spectrometry (ICP-MS). Through ANOVA and PCA, the contents of Cd, Cr, As, Hg, Se, Co, Ni, Sr and Fe mineral elements and the values of $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$ were selected as the variables for the linear discriminant model. Finally, the accuracy of origin discrimination reached more than 95% by leave-one-out cross validation method (Fan et al., 2018). Wang et al. (2020) analyzed the Semen Cassiae from different origins, it was found that the accuracy of origin identification determined by mineral element content (87.1%) was significantly higher than that of stable isotope (51.6%), while the accuracy of mineral element combined with stable isotope was significantly improved (93.5%).

5 Conclusion and prospect

With the diversified development of varieties, planting methods and growth environment of Chinese medicinal materials, the origin traceability of Chinese medicinal materials will be more and more difficult. The two analytical methods have their own advantages: the stable isotopes carry the information of environmental factors, which can reflect the external environmental factors of biological growth, and have high stability that are not easily affected by factors such as processing technology. Mineral elements have geologically specific, and the mineral element analysis method can clarify the characteristics of mineral elements in Chinese medicinal materials, so as to explore the relationship with soil elements. From the existing research, the accuracy of most Chinese herbal medicine origin discrimination models built by mineral elements analysis is higher than that of stable isotope analysis based, and the effect of stable isotope combined with mineral element analysis is better than that of single analysis method. In terms of research content, the existing research objects of origin traceability of Chinese medicinal materials are mainly plant Chinese medicinal materials, and there are few studies on minerals and animals. In terms of data analysis, CA, PCA and LDA algorithms are mainly used currently. Some network neural algorithms, such as decision tree and SVM are also gradually applied to the origin traceability of Chinese medicinal materials. On the other hand, the origin traceability research also tends to multi-element joint chemometric analysis from single element index analysis and simple regional difference analysis. The research of isotope technology and mineral element technology in the origin traceability of Chinese medicinal materials started relatively late, and there are few reports in this regard. Most Chinese medicinal materials are still in the exploratory stage of effective traceability indicators, and no complete database has been established for practical traceability. At present, most of the effective traceability indexes of Chinese herbal medicines are still in the exploratory stage, and a complete database has not been established for practical traceability. In the future, it is necessary to conduct in-depth research from the following aspects.

1) In-depth studies for more isotope application indicators. At present, the studies focus is on the isotopes of C, H, O, N etc., and other isotopes is less, but these isotopes may carry more information about the genuineness of Chinese herbal medicines;
2) It is necessary to expand the geographical scope and quantity of sample collection, screen and expand the traceability index of the regional characteristics, establish and improve the traceability and identification model of various types of Chinese medicinal materials based on chemometrics analysis, and provide an easy-to-operate traceability platform for genuine medicinal materials;
3) It is necessary to study the isotope fractionation mechanism of Chinese medicinal materials in different regions, further explore the influence of climate, topography and geology on the isotope and mineral element composition of Chinese medicinal materials, and make a scientific and reasonable explanation for the genuineness of Chinese medicinal materials combined with the determination of effective components in Chinese medicinal materials. Maybe, it will provide the feasible supports for the cultivation in different places;
4) On the basis of a large number of samples, combined with other technical means, such as spectroscopy, chromatography, etc., to determine the chemical composition fingerprints of traditional Chinese medicines in different regions, supplement and enrich the origin information, and improve the model discrimination accuracy;
5) Establish a standard system for traceability of traditional Chinese medicinal materials, check from the source, involve all links of medicinal materials from planting, harvesting, processing to market circulation, and regulate the adulteration and mixed use of authentic medicinal materials in the market.

Acknowledgements

This work was supported by the National Key R & D Plan (2020YFC1712700) and the Doctoral Research Fund of Guizhou Normal University, China (GZNUD [2019]6).

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