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Geographic Determination of Sangyod Rice using Elemental Concentration and Stable Isotopes Combined with Linear Discriminant Analysis

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Rice is a main food for Thai population and the major cash crop of Thailand. Sangyod rice is traditional rice variety of southern Thailand mainly grown in Phatthalung province and was also registered as the Geographical Indication (GI) and the European Protected Geographical Indication (PGI). The traceability of rice geographical origin has become the most important issue in safety, rice quality assurance, and consumer confidence from mislabeling and adulteration. Stable isotope analysis is a powerful technique for food traceability. This study aimed to discriminate rice origin between Phatthalung and Songkhla provinces that are adjacent provinces. The stable isotopes and elemental concentrations combined with linear discriminant analysis (LDA) and radar plot were also investigated. As the result, three variables (%C, δ^{18}O and %O) were chemical indicators applied to discriminate the geographical origin of Sangyod rice grown in 2 contiguous provinces (Phatthalung and Songkhla). The classification of Sangyod rice origins was correctly accomplished 95.3% of their original groups and 92.1% of cross-validation.

Keywords: Sangyod rice; stable isotope; elements; geographical origin; LDA

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Rice is a staple food for over half of the world's population and 90% of the rice consumption is in Asia including Thailand [1]. Rice is considered the main cash crop of Thailand for export. Thailand is the world’s second largest rice exporter after India [2]. There are many rice varieties in Thailand. Sangyod rice is a local rice variety produced in the southern part of Thailand [3]. Sangyod rice is mainly grown in Phatthalung province that can be grown once a year. It is a distinctive characteristic, small slender grains, soft and pleasant fragrance [4]. The seed coat is red to dark red or pink hue and rich with nutrients and antioxidants, helping to slow down aging and nourishing the blood, preventing memory loss and reduce the risk of cancer [5-6]. Sangyod rice is the first rice variety which has been registered as a geographical indication (GI) product of Thailand on June 23, 2006 under the name of “SANYOD MUANG PHATTHALUNG”. It has been also registered as the European Protected Geographical Indication (PGI) on October 12, 2016 [3,7-8]. Sangyod rice is a high-value product and has more higher prices than other red rice. As a result, it may be the target for adulteration and mislabeling. Therefore, the verification of geographical origins of rice has become a crucial issue to prevent adulteration and mislabeling of rice.

Various analytical techniques such as inductively coupled plasma optical emission spectrometry (ICP-OES), inductively coupled plasma mass spectrometry (ICP-MS) [9], instrumental neutron activation analysis (INAA) [10], laser ablation inductively coupled plasma mass spectrometry (LA-ICP-MS) [11], Fourier transform infrared spectroscopy (FTIR) [12], energy dispersive X-ray fluorescence (ED-XRF) [13], nuclear magnetic resonance (NMR) [14], and elemental analyzer-isotope ratio mass spectrometry (EA-IRMS) [15] have been applied to determine the geographical origins of food products. Stable isotopes have been widely used in the verification of food origins for more than 20 years [16]. Recently, isotopic composition (C, N, O) and elements in foods could be used as indicators to trace their geographical origins of a wide variety of food products i.e., honey [17], beef [18], milk [19], butter [20], virgin olive oil [21], and rice [1,10-11]. Rice is C3 photosynthetic plant that fix carbon dioxide by Calvin cycle [22]. A range of stable carbon isotopic composition (δ^{13}C) values in C3 plants are different from C4 or CAM (Crassulacean Acid Metabolism) plants. The C3 cycle provides δ^{13}C values between −22‰ and −32‰ while C4 cycle has a range of δ^{13}C values between −8‰ and −20‰. [23-24]. In addition to type of plants, the variations of δ^{13}C values depend on the climate and environmental conditions (temperature, relative humidity, light intensity and drought stress) in which the plant grown resulting in different fractionation of CO_2 [22, 24-26]. Nitrogen isotopic composition (δ^{15}N) in plants depends on soil nutrient, agricultural practice, nitrogen fixation, soil moisture, and water irrigation [27-28]. The δ^{15}N values of chemical or synthetic fertilizers for

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conventional cultivation are in the range of -6‰ to 6‰, whereas organic fertilizers have higher $\delta^{15}N$ values (ranges from 5-37‰) than that of conventional cultivation because of the changes of chemical form (nitrogen transformation) in organic matter during the compost fertilizer manufacturing process [28]. Oxygen isotopic ($\delta^{18}O$) fractionation is related to the topography and climate conditions (i.e., altitude, latitudes, longitudes, distance from the coast, the change of evaporation and condensation, precipitation, rainwater, etc.) [29]. Over the past few decades, many researches for determining the geographical origin of rice samples using stable isotope analysis and elemental analysis have been reported. For example, the analysis of C, N, O and S isotope ratios combined with chemometrics has been employed to discriminate rice geographical origin using IRMS analysis [29]. Kelly et al. determined stable isotopes and multi-element in rice samples cultivated in the USA, Europe and Basmati regions using IRMS and ICP-MS. The result revealed that boron, magnesium, carbon and oxygen isotopic composition were used as chemical indicators to distinguish long-grain rice from each country [26]. Suzuki et al. studied carbon and nitrogen contents (C and N contents) and stable carbon, nitrogen, and oxygen isotopic compositions ($\delta^{13}C$, $\delta^{15}N$, and $\delta^{18}O$) of polished Koshihikari rice cultivated in Australia, Japan, and USA using elemental analyzer/isotope ratio mass spectrometry (EA/IRMS) [27].

The determination of geographical origins of rice is difficult when the cultivation areas are close to each other. Therefore, the purpose of this study was to determine the geographical origin of Sangyod rice cultivated in 2 contiguous provinces (Phatthalung and Songkhla) based on the stable isotopes ($\delta^{13}C$, $\delta^{15}N$, and $\delta^{18}O$) and C, N and O contents using EA-IRMS combined with LDA for the first time. Our preliminary results of stable isotopes and element content in Sangyod rice cultivated in the south of Thailand may be valuable database for the authentication or mislabeling of Sangyod rice product.

EXPERIMENTAL

Sample Collection and Preparation

Sangyod rice panicles at maturity were collected from 127 rice samples in 2 provinces (Phatthalung and Songkhla). Paddy rice samples were sun-dried and dehusked by rice milling machine. The unpolished rice samples were pulverized to obtain fine powder by Perten Laboratory mill 3100 (Perten Instruments, Sweden). Then, the fine powder was passed through a 60-mesh (<250 µm particle size) sieve and sealed in cleaned plastic bags. The powdered rice samples were dried in a hot air oven (Memmert, model UN160, Germany) at 60 °C for 48 h until constant weight and kept in a desiccator prior to analysis.

EA-IRMS Analysis

Elemental contents (C, N and O) and stable isotopes of $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$ were analyzed by an Vario PYRO cube elemental analyzer linked to an isotope ratio mass spectrometry (EA-IRMS, Isoprime Ltd, UK). For the $\delta^{13}C$ and $\delta^{15}N$ analysis, powdered rice samples (4 mg) were weighed and then packed into tin boats (4 mm x 4 mm x 11 mm). The encapsulated samples were introduced by autosampler and combusted at 950 °C in a combustion tube. Nitrogen and carbon gases were released and transferred into a reduction tube at 600 ºC while the NO_2 gas was reduced to N_2 gas. The volatile halogen compounds were attached with silver wool in the reduction tube. The reduced gases were passed through a drying tube. Carbon dioxide was adsorbed in an adsorption tube. The adsorption tube was then heated at 240 ºC to release CO_2 prior to IRMS analysis. For $\delta^{18}O$ analysis, the powder rice samples (0.3 mg) were accurately weighed into silver boats (4 mm x 4 mm x 11 mm). The sample boats were folded and then compressed in order to reduce the amount of oxygen. The encapsulated boats were pyrolyzed at 1450 ºC in a glassy carbon reactor, converted to CO gas and transferred by carrier gas into the adsorption tube. The CO gas was separated from N_2 gas in adsorption column and then passed through IRMS system.

The δ (per mil, ‰) notation is used to explain the difference in isotopic composition of rice samples relative to the international reference standards (Vienna Pee Dee Belmnite for $\delta^{13}C$, Air for $\delta^{15}N$, and Vienna Standard Mean Ocean Water, VSMOW for $\delta^{18}O$) defined as the following equation 1:

$$\delta\ (‰) = \frac{(R_{sample} - R_{standard})}{R_{standard}} \times 1000 \qquad (1)$$

where R_{sample} and $R_{standard}$ are the isotope ratio of the sample and isotope ratio of the international standard defined by IAEA (International Atomic Energy Agency), respectively.

Statistical Analysis

The multivariate analysis of the obtained data from EA-IRMS was performed by IBM SPSS statistics 23 software. The LDA based on stable isotopes and elements were applied to discriminate the geographical origin of Sangyod rice grown in 2 contiguous provinces (Songkhla and Phatthalung).

Table 1. The percentage of C, N and O in Sangyod rice samples cultivated in Songkhla and Phatthalung.

Province	Elemental content (%)					
	C		N		O	
	Mean	S.D.	Mean	S.D.	Mean	S.D.
Songkhla	41.33	0.56	1.47	0.18	42.27	0.83
Phatthalung	42.70	0.61	1.43	0.11	42.57	0.81

RESULTS AND DISCUSSION

Element Contents in Sangyod Rice by Elemental Analyzer

C, N, and O contents in Sangyod rice cultivated in Songkhla and Phatthalung provinces were determined. C contents cultivated in Songkhla and Phatthalung provinces were found in the ranges of 40.12% to 42.46% and 41.57% to 44.15%, respectively. N contents in Sangyod rice grown in Songkhla and Phatthalung provinces were 1.10% to 2.01% and 1.20% to 1.84%, respectively. O contents in Sangyod rice cultivated in Songkhla and Phatthalung provinces were in the ranges of 40.54% to 44.05% and 40.82% and 44.63%, respectively. The highest mean concentrations of C and O were found in Sangyod rice obtained from Phatthalung province and the highest mean concentration of N was found in Sangyod rice grown in Songkhla province.

The mean values and standard deviation (S.D.) of %C, %N and %O in Sangyod rice samples cultivated in Songkhla and Phatthalung provinces are summarized in Table 1.

$\delta^{13}C$, $\delta^{15}N$, and $\delta^{18}O$ Analysis

In this study, the stable isotopes ($\delta^{13}C$, $\delta^{15}N$, and $\delta^{18}O$) in Sangyod rice samples cultivated in Phatthalung and Songkhla provinces were determined by EA-IRMS. Figure 1 shows the variable distribution of $\delta^{13}C$, $\delta^{15}N$, and $\delta^{18}O$ in 2 contiguous provinces. The $\delta^{13}C$ values of Sangyod rice grown in Songkhla and Phatthalung provinces were in the ranges of -29.86‰ to -27.40‰ and -28.90‰ to -27.25‰ with the mean values of -28.40 and -28.26‰, respectively.

The $\delta^{15}N$ values found in Sangyod rice obtained from Songkhla and Phatthalung provinces were in the ranges of +0.50‰ to +9.28‰ and +1.05‰ to +8.48‰ with the mean values of +4.26 and +4.34‰, respectively. The distributions of $\delta^{13}C$ and $\delta^{15}N$ values in Sangyod rice cultivated in Songkhla and Phatthalung provinces are shown in Figure 1.

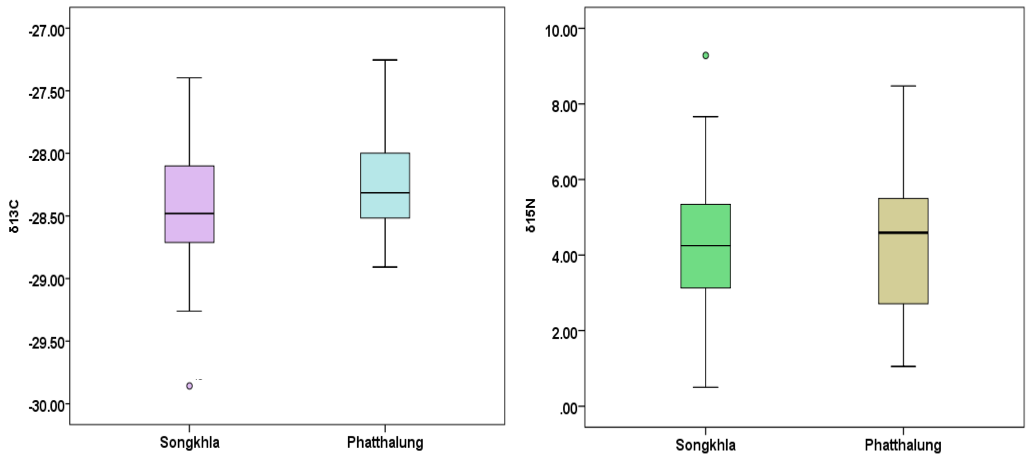

Figure 1. Box plots showing the distributions of $\delta^{13}C$ and $\delta^{15}N$ values in Sangyod rice cultivated in Songkhla and Phatthalung provinces.

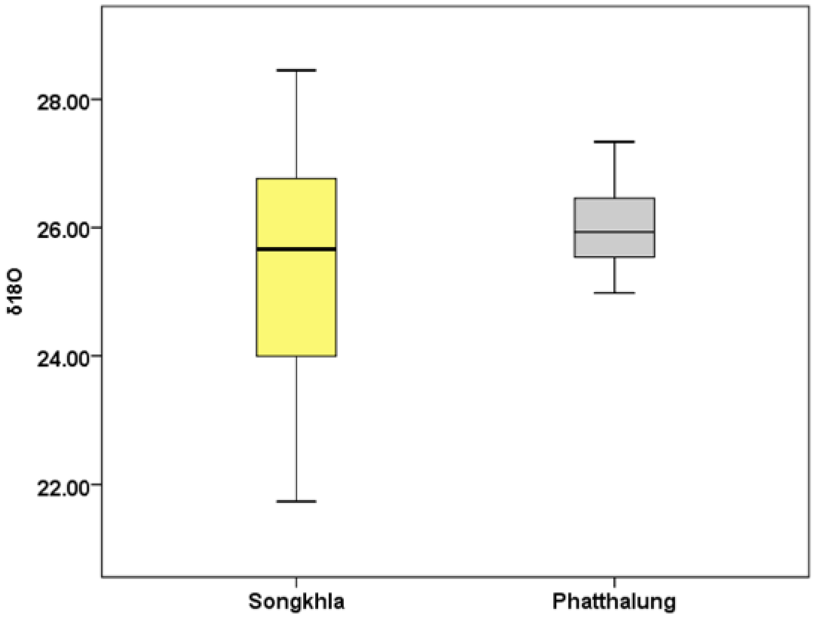

Figure 2. Box plots showing the distributions of $\delta^{18}O$ values in Sangyod rice cultivated in Songkhla and Phatthalung provinces.

Table 2. Isotopic compositions of C, N and O in Sangyod rice samples cultivated in Songkhla and Phatthalung provinces.

Province	Isotopic composition (‰)					
	$\delta^{13}C$		$\delta^{15}N$		$\delta^{18}O$	
	Mean	S.D.	Mean	S.D.	Mean	S.D.
Songkhla	-28.40	0.46	+4.26	1.62	+25.40	1.68
Phatthalung	-28.26	0.36	+4.34	1.82	+25.73	1.30

The $\delta^{18}O$ values of Sangyod rice cultivated in Songkhla and Phatthalung provinces ranged from +21.74‰ to +28.45‰ and +24.98‰ to +27.33‰, respectively. The mean $\delta^{18}O$ values in Sangyod rice obtained from Songkhla and Phatthalung provinces were +25.40‰ and +26.05‰, respectively. The distributions of $\delta^{18}O$ values of Sangyod rice cultivated in Songkhla and Phatthalung provinces are shown in Figure 2.

The highest mean values of $\delta^{13}C$, $\delta^{15}N$, and $\delta^{18}O$ were found in Sangyod rice cultivated from Phatthalung provinces. The mean concentrations of $\delta^{13}C$, $\delta^{15}N$, and $\delta^{18}O$ in Sangyod rice samples cultivated in Songkhla and Phatthalung provinces are summarized in Table 2.

Radar Plot Analysis

Elemental contents and isotopic compositions in Sangyod rice cultivated from the 2 contiguous provinces were plotted by radar plot analysis. The radar plot based on mean values of %C, %N, %O, $\delta^{13}C$, $\delta^{15}N$, and $\delta^{18}O$ is illustrated in Figure 3. From the radar plot analysis, it was quite similar pattern of the studied variables in Sangyod rice cultivated between Songkhla and Phatthalung provinces because Sangyod rice is usually cultivated using the same condition. As a result, it is difficult to discriminate the geographical origin of Sangyod rice cultivated between 2 contiguous provinces by radar plot analysis as shown in Figure 3.

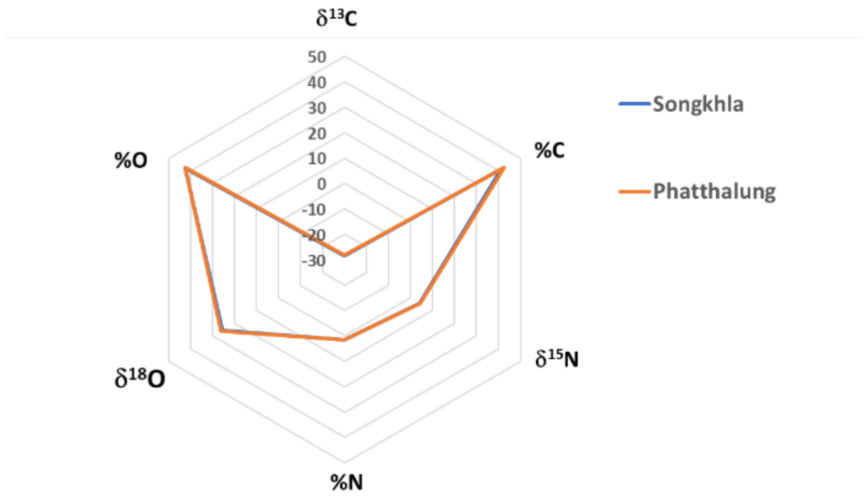

Figure 3. Radar plot illustrating the variables in mean values of %C, %N, %O, $\delta^{13}C$, $\delta^{15}N$, and $\delta^{18}O$ in Sangyod rice cultivated in Songkhla and Phatthalung provinces.

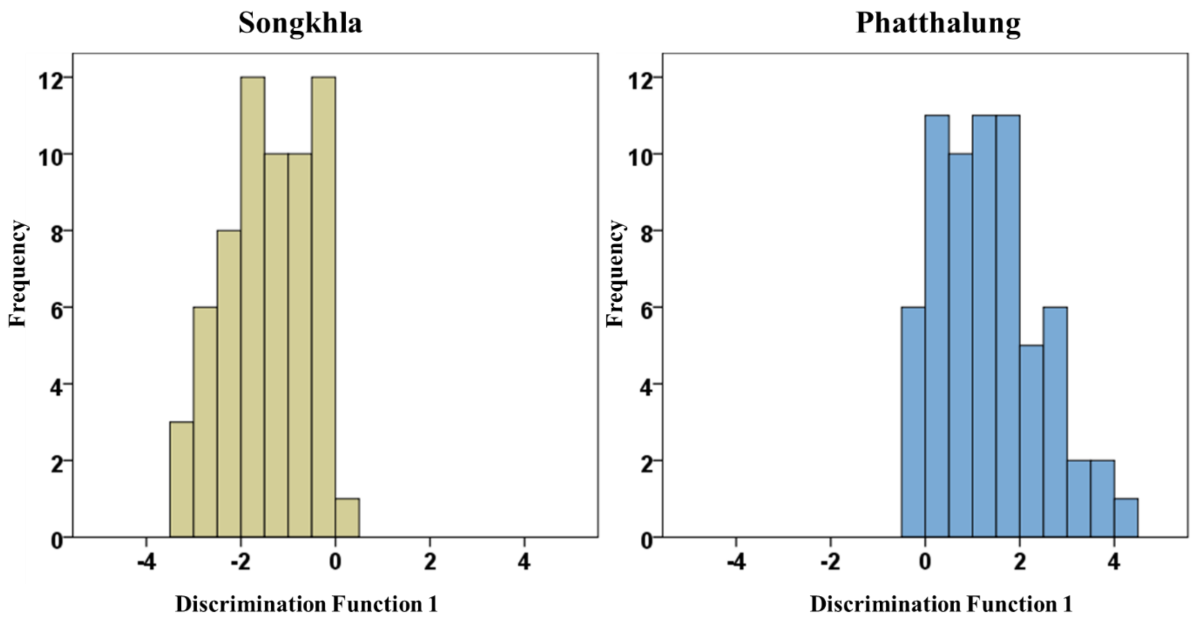

Figure 4. LDA result based on 6 variables (%C, %N, %O $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$) in Sangyod rice cultivated in Songkhla and Phatthalung provinces.

LDA

In order to determine the geographical origin of Sangyod rice cultivated in 2 contiguous provinces, the variables including %C, %N, %O, $\delta^{13}C$, $\delta^{15}N$, and $\delta^{18}O$ in Sangyod rice samples cultivated from Songkhla and Phatthalung provinces were combined with LDA. The result of LDA based on the six variables for determining the geographical origin of Sangyod rice cultivated in two contiguous provinces is shown in Figure 4. There were significant differences in %C, %O and $\delta^{18}O$ values with the p-value < 0.001, 0.037 and 0.005, respectively.

Table 3. Classification of the geographical origin of Sangyod rice cultivated in Songkhla and Phatthalung provinces.

Origin			Predicted group membership		
			Songkhla	Phatthalung	Total
Original	count	Songkhla	61	1	62
		Phatthalung	5	60	65
	%	Songkhla	98.4	1.6	100.0
		Phatthalung	7.7	92.3	100.0
Cross-validated	count	Songkhla	58	4	62
		Phatthalung	6	59	65
	%	Songkhla	93.5	6.5	100.0
		Phatthalung	9.2	90.8	100.0

Figure 4 presents the distributions of Sangyod rice according to the geographical origins (Songkhla and Phatthalung provinces) using %C, %N, %O, $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$ values. The correlation of discriminant function 1 based on six variables (%C, %N, %O, $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$) was 0.809. Function 1 explains 100% of the total variance. Function 1 was related to %C with a standardized canonical discriminant function coefficient of 1.110. Therefore, LDA combined with the 6 variables (%C, %N, %O, $\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$) showed good accuracy with 95.3% correct classification of their original groups and 92.1% of cross-validation as shown in Table 3. The results demonstrated that the geographical origin of Sangyod rice samples was successfully separated from 2 contiguous provinces using the combination of the element contents (%C, %N and %O), isotopic compositions ($\delta^{13}C$, $\delta^{15}N$ and $\delta^{18}O$) and LDA.

CONCLUSION

The discrimination of Sangyod rice cultivated in Songkhla and Phatthalung provinces was successfully implemented by elemental contents and stable isotopes combined with statistical analysis. The result indicated that three variables (%C, $\delta^{18}O$ and %O) showed to be of high potential as chemical indicators to discriminate the geographical origin of Sangyod rice grown in 2 contiguous provinces (Phatthalung and Songkhla). There were no significant differences in $\delta^{13}C$, %N and $\delta^{15}N$ with the p-value > 0.05 between Sangyod rice cultivated in Phatthalung and Songkhla provinces. LDA provided good accuracy with correct classification and cross-validation of 95.3% and 92.1%, respectively. Our results may provide the preliminary information for discrimination the geographical origin of Sangyod rice samples in 2 contiguous provinces (Phatthalung and Songkhla) and it is also possible to trace the geographical origin of rice cultivated from their different origins.

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