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REVIEW

Authenticity of the Geographical Origin and Production Methods of Agricultural Products

– Application of Element Composition and Stable Isotope Analyses –

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Abstract

Multi-element analysis including stable isotopes can be used as a possible indicator for food safety and security. For the certification of geographical origin, the analytical methods can be performed in two ways: one where multivariate analysis is used to determine the concentrations of such omnipresent elements as Al, Ca, Cl, Mg, Mn, Fe and Zn, and one that focuses on such special elements as the stable isotope ratios of Sr, O, and H. For the certification of production methods, especially those regarding organic products, δ^{15}N values could be a potential indicator, particularly in such protected cultivations as in a plant factory (advanced-type greenhouse horticulture). Because the accuracy of these values is affected by production conditions, the δ^{15}N values of products can be predicted more accurately under controlled conditions, such as in a plant factory using δ^{15}N-evaluated fertilizer, medium, and water. Non-destructive systems have been developed for measuring both the level of elements in a product and the production environment, such as soil conditions. In the near future, the results of chemically analyzed and those of non-destructively analyzed elemental composition will become interconnected to non-destructively certify the geographical origin and production method of agricultural products. All these destructive methods have been used to a limited extent for practical regulation as an analysis guideline; however, a combined system involving the use of the newly developed detector (non-destructive), data collection, and analysis using artificial intelligence could address the issue of falsely labeled products for practical application. Particularly in a plant factory, production is controlled and regulated, allowing the tracing of products from the farm to the table. In this context, the greenhouse production system would be an advanced example for the practical use of food safety combined with analytical chemistry and information communication technology.

Discipline: Agricultural environment
Additional key words: agricultural product, authenticity, geographical origin traceability, element composition, stable isotope

Diversity of agricultural products and food caused by enhanced trade and complex production methods

1. Social background

There has been growing concern about food security in recent years. The import and export of agricultural products could increase in the coming decades. And the Japanese government plans to set numerical targets for agricultural and food exports. It aims for producers to ship products worth one-trillion yen (or about eight-billion dollars) in 2020 (Honda 2014).

In the future, organic products and organic fertilizer may become more strictly regulated worldwide due to the growing concern of consumers regarding food safety, and as organic products are generally considered safe food. Despite these situations, a Japanese agricultural federation (Zen-Noh) has recalled 10,000 tons of falsely labeled organic fertilizers, the products obtained from which were thought to be safe to use, but contained falsely labeled organic contents (Zen-Noh 2015). In considering existing international and complex production systems, a method of standardization for food reliability that is suitable for practi-

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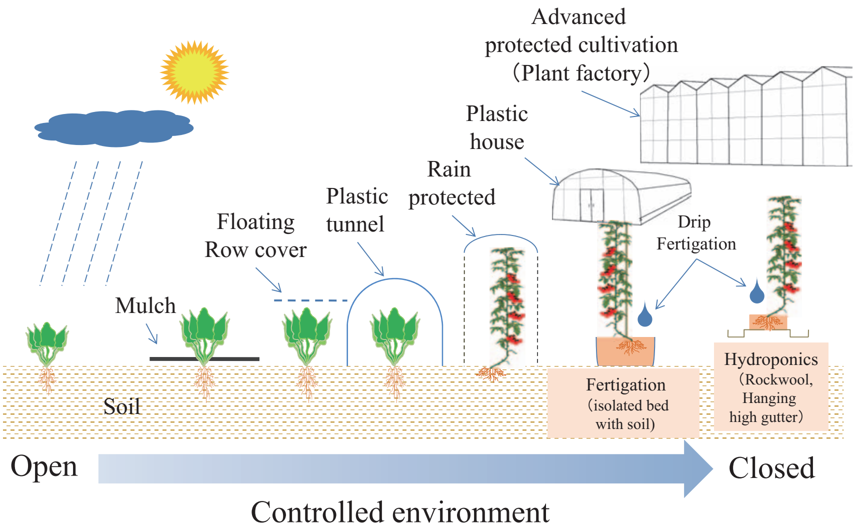

Fig. 1. Various production systems for agricultural products

cal use in terms of food safety and security needs to be established immediately.

2. Which country and which agricultural production system

In developed countries, many consumers believe that the production systems in their countries are sophisticated, although this is not necessarily true. Thus, the country of production is one of the concerns regarding consumption, although the actual situation of agriculture production is rather complex. There are many kinds of production systems, ranging from open to closed ones (Fig. 1). In each system, fertilization and chemical spraying regimens differ, and can be used in numerous combinations. Therefore, some standardized indicator needs to be developed. Whether a production system involves the use of chemicals is of primary concern, as consumers typically consider organic products to be safe. However, these requirements could vary across countries; in general, synthetic chemical inputs (e.g., prohibited fertilizers, and pesticides) have been avoided in the fields for many years (often, three or more). The commercial use of the term "organic" is legally restricted. For production systems, the identification of organic products would have top priority.

Certification of origin and production system of agricultural products and food by chemical analysis

As mentioned above, many consumers are concerned about production processes relative to how agricultural products and food are developed in a country. There are many kinds of production systems, including organic farming. In addition to determining the quality of products, the reliability of their labeling must be assured for selecting the products. Chemical analysis is currently the most suitable method of identifying agricultural products. We have reviewed articles on the application of chemical analysis for the identification of agricultural products.

Identification of geographical origin

Recently, many studies have focused on the use of natural abundance of isotope variation and elemental concentrations as geographic tracers to determine the provenance of food products (Kelly et al. 2005). These investigations exploit the systematic global variations of stable hydrogen and oxygen isotope ratios in combination with elemental concentrations, including heavy isotope variations (e.g., Sr). Some examples of applying the multi-isotopic and multi-element analysis methods of food forensics are summarized below.

1. Analysis of mineral composition for the determination of geographic origin

The mineral composition of taro (*Colocasia esculenta* (L.) Schott) was analyzed to develop a method of distinguishing taro produced in Japan and that produced in China (Kobayashi et al. 2011). The concentrations of 21 elements were assayed using instrumental neutron activation analysis or ion chromatography. The mean concentrations of $H_2PO_4^-$, Co, Cr, and Na significantly differed between the taro grown in Japan and that grown in China. The highest percentage of correct classification was achieved with a two-variable model that included $H_2PO_4^-$ and Co.

A chemometric study was conducted to identify the provenance of a specific agricultural product (beef). Instrumental neutron activation analysis and prompt gamma-

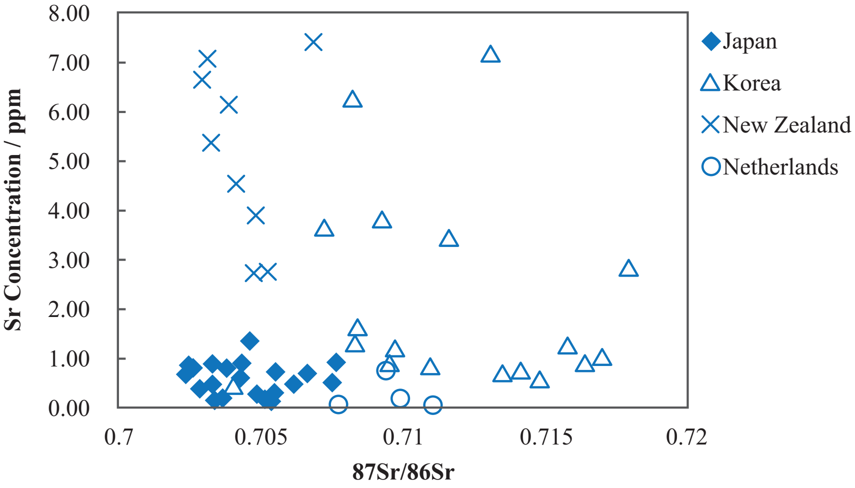

Fig. 2. Sr concentration versus Sr isotope ratio of samples originating from Japan (Tsukuba and Mito), Korea, New Zealand, and the Netherlands

ray analysis were used in combination to non-destructively determine the concentrations of 10 elements—C, H, N, S, Sm, Br, Mg, Na, K, and Cl (Saito et al. 2008). The elemental profiles of beef samples produced from different regions in Japan, Australia, and the USA were determined. The Holstein beef of Japan and that of Australia were not properly grouped by principal component analysis modeling using the elemental data set. This is a representative example showing that such an approach does not always yield successful results, and that a trial-and-error approach is necessary.

Some of the many examples are described below. In general, the successful identification of geographical origin by using multivariate analysis and identification of the concentrations of omnipresent elements have been reported in onion (Ariyama et al. 2007), welsh onions (Ariyama et al. 2004), spinach (Yanada et al. 2007), taro (Nakamura et al. 2012), and tea (Pilgrim et al. 2010).

2. Sr stable isotope ratio to identify the geographical origin

The accuracy of multivariate analysis with inorganic element composition analysis to detect the provenance of food products might be improved by using the Sr isotope ratio ($^{87}Sr/^{86}Sr$). The Sr isotope may be a strong tool for food forensics. The water or soil Sr isotope ratio could affect the values of products. The Sr isotope ratios of about 650 different European natural mineral waters were investigated (Voerkelius et al. 2010). The analyzed $^{87}Sr/^{86}Sr$ values in different waters ranged from 0.7035 to 0.7777, which indicated that these values are influenced by the remarkable diversity of rocks from young mantle-derived basaltic rocks to very old silicic continental crust. The resulting map can be used to predict the Sr isotopic composition of plants grown in different waters. Such a chemical analytical approach could provide a cost-effective and preliminary screening tool.

The Sr isotope ratio of brown rice (*Oryza sativa* L.) was determined using multicollector-inductively coupled plasma mass spectrometry (MC-ICP-MS) to estimate the production area of rice (Kawasaki et al. 2002). The Sr isotope ratios of Japanese rice samples ranged from 0.706 to 0.709, which were slightly lower than those of Chinese and Vietnamese rice samples (0.710 to 0.711). Australian rice showed the highest Sr isotope ratio (0.715 to 0.717); in contrast, Californian rice (0.706) showed a ratio lower than those of almost all the Japanese samples. These results suggested that the Sr isotope ratios could provide key information for estimating the geographical origin of rice.

The Sr stable isotope ratio was also applied to identify the geographical origin of paprika. The Sr isotope ratio and trace elements (Al, Ti, Mn, Fe, Ni, Cu, Zn, Rb, and Sr) of 52 paprika samples from Japan, Korea, New Zealand, and the Netherlands were measured (Fig. 2; Tsuchida et al. 2014) by using double-focusing inductively coupled mass spectrometry. The Sr isotope ratio combined with trace element signature was found to be suitable for estimating the production sites of paprika.

Another example concerns processed food; a suitable method of determining the Sr isotope ratio in wines by using ICP-MS has been developed and applied to ten samples of table and fortified wines from five different regions of Portugal and one region in France (Almeida et al. 2001).

3. O, H, C, and N stable isotope ratios to identify the geographical origin of agricultural products

Among the rice from 14 different cultivation areas, including Australia, Japan, and the USA, the cultivated area

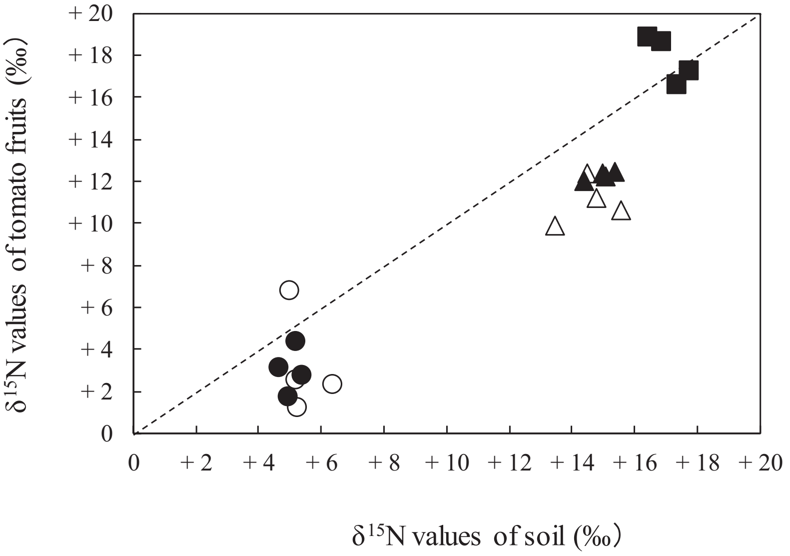

Fig. 3. Relationship between δ^{15}N values of soil and tomato fruits
CDU: ●, compound fertilizer; LSR: ○, low-sulfate slow-release fertilizer
CM + CDU: ▲; PM + CDU: △; CM + PM: ■
CM: cattle manure, PM: poultry manure

was clearly distinguished by a pentagonal radar plot developed based on the elemental and isotopic compositions (Suzuki et al. 2008). These results suggested that a comparison of C and N contents and the δ^{13}C, δ^{15}N, and δ^{18}O values could potentially be useful for the rapid and routine discrimination of the geographical origin of polished rice.

The stable isotope ratios of C, O, and H and their elemental composition were also used to check the authenticity of Italian extra-virgin olive oils (Camin et al. 2010), apple (Suzuki et al. 2012), and fruit juices (Magdas & Puscas 2011).

A comparison of paprika fruits with different geographical origins (Japan, Korea, and the Netherlands; Nakano et al. 2010) showed δ^{15}N of +6.0% for Japanese fruits compared to +0.2% for Korean fruits and +3.1% for Dutch fruits. These differences suggested that paprika was cultivated using different cultivation and fertilizer application methods in the three countries. Lower δ^{15}N of a product suggests that it was produced by hydroponics (without soil), as soil nitrogen showed higher δ^{15}N. These data also suggest that this method can be used to identify the geographical origin of agricultural products.

Identification of organic products

The δ^{15}N characteristics of agricultural products cultivated using different fertilization regimes have been reported in many crops. Many studies have mainly focused on organic products (Georgi et al. 2005, Rapisarda et al. 2005, Flores et al. 2007). In this section, we summarize the results of studies that analyzed the δ^{15}N values for a particular crop and yielded consistent results by conducting many experiments.

1. Leafy vegetables

The δ^{15}N values of leaf mustard were correlated with those of the applied chemical, organic, and mixed fertilizers (Nakano & Uehara 2006a). A linear regression of δ^{15}N values of fertilizer and leaf mustard (*Brassica campestris* L.) yielded R = 0.88. These results suggested that a database of δ^{15}N values of fertilizers could be useful for the scientific certification of products.

2. Fruits and vegetables

We determined the effects of chemical fertilizer and compost application on the yield and quality of tomato (*Lycopersicon esculentum* Mill. ‘Saturn’) in an isolated bed (Nakano et al. 2003). Five treatments were conducted within a two-year period of 4 continuous croppings system. Compound fertilizer (CDU) and low-sulfate slow-release fertilizer (LSR) were used for the chemical fertilizer plots (Fig. 3). A mixture of cattle manure (CM) and CDU (CM + CDU), a mixture of poultry manure (PM) and CDU (PM + CDU), and a mixture of cattle and poultry manure (PM + CM) plots were arranged as compost-used plots. We then measured the δ^{15}N values of tomatoes and those of the soil for each treatment, and then estimated the correlation of δ^{15}N values between the fruits and soil to certify the compost-applied products.

We did not detect any differences in the yield and sugar content among the treatments. These results showed

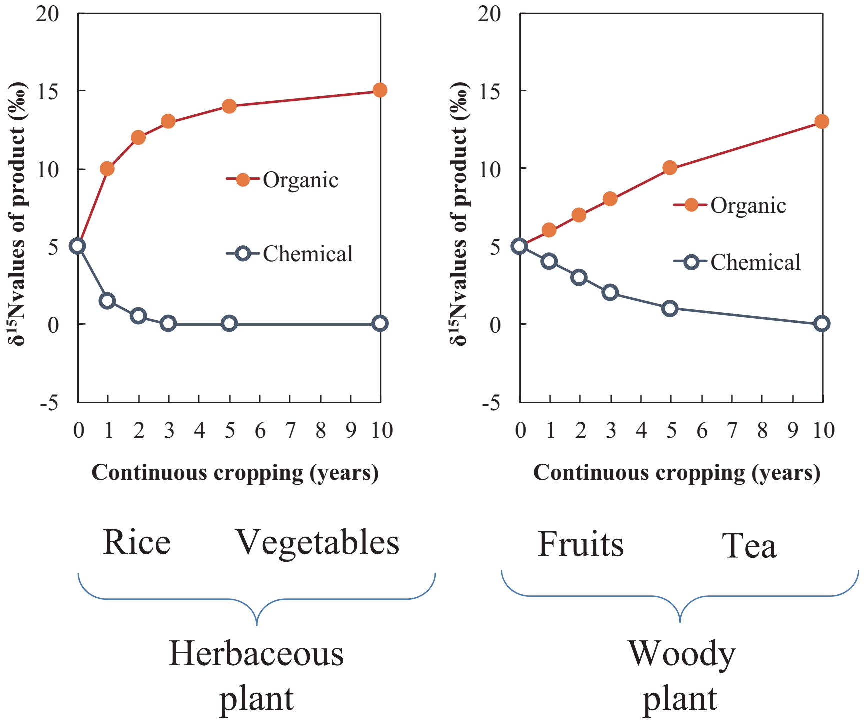

Fig. 4. Changes in δ^{15}N values of herbaceous and woody plant products over time

that conducting organic fertilizer application is not always beneficial for tomato yield and quality. Conversely, the δ^{15}N values of tomato fruits showed significant differences among fertilizer applications. The δ^{15}N values of chemical fertilizer application were –1.6% and –1.1% for CDU and LSR, respectively, and those for the mixture of chemical and compost were +14.1% and +14.7% for CM + CDU and PM + CDU, respectively. The mixture of PM and CM showed the highest δ^{15}N values (+17.9‰) among the treatments. During the composting process, the δ^{15}N values of the compost increased to +17.2% within 30 days (Nakano & Uehara 2009). In that study, nitrogen isotope discrimination by ammonium volatilization and denitrification within 30 days after composting commencement mainly determined the δ^{15}N values of compost.

The δ^{15}N values of the soil and fruits reflected those of the fertilizers and were positively correlated ($R^2 = 0.89$). The δ^{15}N values could be used as an indicator of organic products by setting a threshold point, such as using a threshold of +5.0% to distinguish products cultivated with chemical and organic fertilizers. And such an analytical approach provides a cost-effective and preliminary screening tool. In the organic fertigation method, organic tomato (Nakano & Uehara 2003) and melon (Nakano & Uehara 2005) showed higher δ^{15}N values than those in the conventional chemical fertigation system. In the market, organic certified vegetables (tomato, cucumber, eggplant, sweet Japanese pepper, and pumpkin) also showed higher δ^{15}N values than the +5.0% threshold (Nakano et al. 2002).

3. Fruits and tea

For fruits, in contrast to leafy vegetables, the applied δ^{15}N values (fertilizer, water, and soil) gradually reflected those of fruits (Saito 2008), such as in tea plants with new leaves (Nakano et al. 2005a). More than three years are necessary to significantly differentiate between products (Fig. 4; Nakano 2010). Given the vast nitrogen sink because of the trunks and roots in tree plants, developed fruits and new leaves would be affected by the long history of the environment (i.e., long-term soil conditions, fertilization).

4. Rice

The characteristics of δ^{15}N values between rice and soil in organic farming and conventional farming were examined, and the possibility of discriminating organically grown rice from conventionally (non-organically) grown rice by using this relationship was explored (Nishida & Sato 2015). The δ^{15}N values of organically grown rice tended to be higher than the regression line obtained from the δ^{15}N values of rice and soil without N source application (consisting of mostly soil and water nitrogen). The δ^{15}N values of conventionally grown rice tended to be lower than these values. The variation of δ^{15}N values was wide, however, and the difference in δ^{15}N values between conventional and organic farming became ambiguous as rice is typically culti-

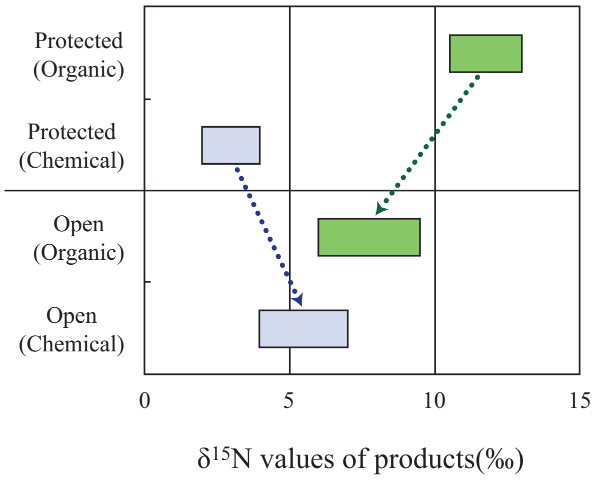

Fig. 5. Effect of cultivation field (open and protected) and chemical/organic fertilization

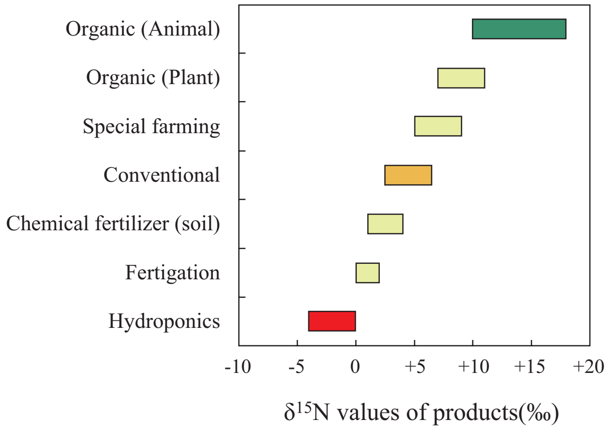

Fig. 6. Difference in δ^{15}N values of products among production systems

Special farming: less than half the amount of chemical fertilizer and less than half the times of agrochemical spraying as compared to conventional farming.

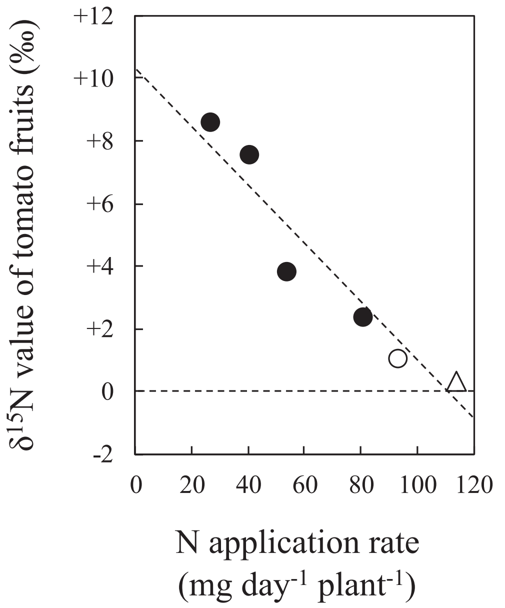

Fig. 7. Effects of N application rate in fertigation on δ^{15}N values of tomato fruits

●: Data on inorganic fertigation from Nakano et al., (2005b); ○: Data on inorganic fertigation from Nakano et al. (2004); and △: Data on inorganic fertigation from Nakano et al. (2003).

vated in open fields affected by rain leaching. These results also indicated that the δ^{15}N values of rice and soil could aid in discriminating between organic and conventional rice.

General conclusion of organic certification using δ^{15}N values and their relation to production systems

1. Production systems and δ^{15}N—open field or protected cultivation

The δ^{15}N values of organic rice, tea, and fruits were relatively lower than those of vegetables grown using protected cultivation. In protected cultivation, the effect of rainfall is limited and little leaching occurs. The difference in the δ^{15}N values of fruits cultivated in open and closed systems can be attributed to the higher dilution effects of fertilizers in the open cultivation system than in the closed system (Fig. 5; Nakano 2010). Taken together, these results suggest that the δ^{15}N values of products differed among production systems (Fig. 6). The δ^{15}N values of products obtained using animal compost were the highest due to the concentrated ^{15}N of compost, and that of hydroponic products were the lowest because chemical fertilizer is synthesized from nitrogen in the air that has a lower ^{15}N concentration than any other nitrogen source.

2. Estimation model for δ^{15}N values of a product

We hypothesized that the δ^{15}N values of products would be reflected by the ratio of he values of tertilizer, soil and water, especially in protected cultivation. Under restricted conditions, the nitrogen source was significantly restricted.

First, fertilizer is known to have a prominent effect on the δ^{15}N values of products. The results of a dose-response experiment conducted using different application rates of chemical fertigation revealed that the greater the application of chemical fertilizer, the higher the dilution of soil nitrogen effects (Fig. 7; Nakano et al. 2005b, Nakano 2010). Second, soil nitrogen has an effect on the δ^{15}N values of products. In cases where no chemical and organic fertilizer are used, the δ^{15}N values of soil reflected those of the products, such as strawberry (Nakano & Uehara 2006b) and tomato (Nakano & Uehara 2008). Finally, water also affects the δ^{15}N values of products, though its effect would be limited. In many cases, the effect of water would be negligible.

Based on these results, we adapted two source models to predict the $\delta^{15}N$ values of the products. If we consider the $\delta^{15}N$ values of fertilizer, soil (PEN: phosphate-extracted nitrogen), and plant (products) as δf, δs, and δp, then their N concentrations would be Nf, Ns, and Np, respectively, which can be calculated as:

$$\delta p = (\delta f \times Nf + \delta s \times Ns)/(Nf + Ns)$$

The $\delta^{15}N$ of products would be expected to show a high correlation to the characteristics of fertilizer and soil (R = 0.893; Fig. 8; Nakano & Uehara 2008).

Food security and protected cultivation

1. Plant factory

Along with the recent advances made in protected cultivation, the number of plant factories has also increased (Fig. 9; Nakano et al. 2012) as more efficient and high-quality production systems must be developed to overcome cheaper imported products. As mentioned before, under the protected condition, connecting a production practice to products is easier. If stable isotopes were used to certify the origin and history of products under this condition, the most advanced secured system would be obtained. Moreover, food poisoning will be minimized due to the use of strictly controlled environments such as that in a plant factory.

2. Secured production of vegetables in Asian countries

For securing the reliability of agricultural products and food, many researchers aim to establish methods that involve the use of such components as heavy and light elements, including stable isotopes and combinations thereof. These scientific evaluation criteria would enable the development of safer diets for people. Moreover, a plant factory might provide the ultimate safe agricultural products in the future, especially vegetables.

Finally, the collaborative findings from Japan, China, Korea, and other southeast Asian countries (ASEAN) would be valuable in providing more safe agricultural products to people in Asia's economic regions. The demand for both safe and secure food is expected to increase in the coming decades. Countries in the Asian region face similar problems, especially regarding food poisoning caused by high temperature and high humidity, and dietary habits (i.e., a rice-based diet).

3. Food safety combining analytical chemistry and information communication technology

All methods for food authenticity developed thus far have no more than one partial use for practical application as an analysis guideline. For both measuring the products and determining the production environment (e.g., soil conditions), a non-destructive system has been developed (Zhao & Nakano 2018). An environmental measurement node using open source hardware (UECS: Ubiquitous environmental control system) has also been developed (Yasuba et al. 2013). These innovative tools could facilitate the practical application of food security.

In the near future, chemically analyzed results and non-destructive analysis results will be connected to certify the geographical origin and production methods used for practical purposes.

A combined system involving the use of the newly developed non-destructive detector could enable data collection and analysis using information communication technology (ICT), including UECS and artificial

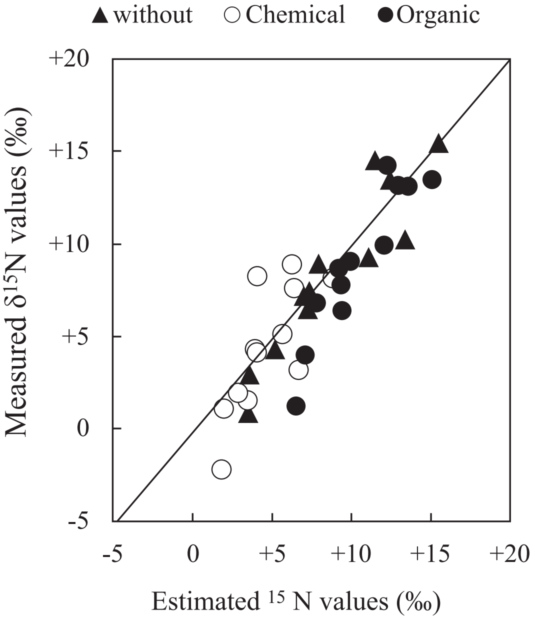

Fig. 8. Relationship between estimated and measured $\delta^{15}N$ values of tomato fruits cultivated using different fertilization regimes
Without fertilization: ▲; Chemical fertilization: ●; Organic fertilization: ○.

Fig. 9. Plant factory at NARO Tsukuba
A high-yield and high-quality production system.

intelligence; consequently, this could address the problem of falsely labeled products. Particularly in a plant factory (advanced-type greenhouse horticulture), production is controlled and regulated, so that products suitable for tracing from the farm to the table are obtained. In this context, greenhouse production systems can be considered for practical application of food safety combined with analytical chemistry and ICT.

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