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REVIEW

Natural ^{15}N Abundance Can Aid the Discrimination of Organic and Conventional Rice

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Abstract

The natural ^{15}N abundance ($\delta^{15}N$ value) of organic rice tends to be higher than that of conventional rice. However, as $\delta^{15}N$ values vary in both organic and conventional rice, it might be difficult to use a particular $\delta^{15}N$ value as an indicator of organic growth conditions. This review describes an approach that was developed at the Tohoku Agricultural Research Center, National Agriculture and Food Research Organization. The relationship between the $\delta^{15}N$ values of rice and those of soil under organic or conventional farming conditions has been investigated. Regardless of the farming method, the $\delta^{15}N$ values of rice reflect those of the soil. The $\delta^{15}N$ values of organically grown rice tend to be higher than the regression line obtained from the $\delta^{15}N$ values of rice and soil without the application of an N fertilizer. The $\delta^{15}N$ values of conventionally grown rice tend to be lower than the regression line. Thus, the relationship between the $\delta^{15}N$ values of rice and soil without an applied N source could be used to differentiate between organic and conventional rice. However, the existence of regional variation in the relationship between the $\delta^{15}N$ values of rice and those of unamended soil can confound the use of this discriminant method. Such variation may occur due to the differences in $\delta^{15}N$ of natural N inputs, and also through ammonia nitrification and subsequent denitrification. Temporal variation can also occur, though the reason for such variation is unknown. When the relationship between the $\delta^{15}N$ values of rice and those of unamended soil is employed to distinguish between organic and conventional rice, regional and temporal variations in that relationship should be taken into account.

Discipline: Soils, fertilizers and plant nutrition
Additional key words: nitrogen source, organic fertilizer, paddy soil, regional variation, synthetic fertilizer

Introduction

There is a growing demand for plant foods cultivated using organic methods. For example, the production of certified organic rice (*Oryza sativa* L.) in 2015 was 8,831 Mg, thus recording an increase of 15% from 2001 (Ministry of Agriculture, Forestry and Fisheries 2017a). However, there is also the growing problem of deliberately mislabeling conventionally grown plants (fruit, vegetables, and grain) as organically grown produce (Ministry of Agriculture, Forestry and Fisheries 2017b). This type of food fraud is becoming an increasingly important social problem as it can endanger both food safety and security. Therefore, preventive measures against food fraud need to be developed to prevent misrepresentation. Considerable scientific efforts are underway to develop objective methods of distinguishing organically grown food from conventionally cultivated crops. With respect to pesticide use, methods of detecting pesticide residues have been developed (Tanizawa et al. 2005, Kobayashi 2009). To identify the fertilizer management regime during cultivation, it may be feasible to use natural ^{15}N abundance ($\delta^{15}N$ value) to discriminate between organically and conventionally grown crops. The $\delta^{15}N$ values of organically grown crops, which are grown without synthetic fertilizer, tend to be higher than those of conventionally grown crops for which synthetic fertilizer has been used; this difference has been reported

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for vegetables (e.g., Nakano et al. 2003, Bateman et al. 2007, Rogers 2008, Šturm et al. 2011), fruit (Camin et al. 2011), and tea (Hayashi et al. 2011). Synthetic fertilizer is chemically manufactured fertilizer, and its N is derived from atmospheric N with a δ^{15}N value of 0‰. These observations suggest a means to discriminate organic crops through use of the δ^{15}N value.

A pioneer study conducted by Fujita et al. (2003) on the δ^{15}N values of organic rice and conventional rice revealed a difference between both types. Suzuki et al. (2009) also found different δ^{15}N values between organic rice and conventional rice. Recently, Nishida and Sato (2015, 2016) developed a new approach at the Tohoku Agricultural Research Center, National Agriculture and Food Research Organization (TARC/NARO) for discriminating between organic rice and conventional rice through use of the relationship of δ^{15}N values between rice and soil. This review examines studies on the δ^{15}N values of organic rice and conventional rice conducted at TARC/NARO, and includes other relevant studies on distinguishing between organic rice and conventional rice.

δ^{15}N values of rice

It was first reported in 1990 that the δ^{15}N values of rice grown without synthetic fertilizer were higher than those grown with synthetic fertilizers (Yoneyama et al. 1990). The δ^{15}N values of paddy soils as well as rice are higher after repeated application of organic materials, such as crop residue, livestock manure and green manure, without synthetic fertilizer than those applied with synthetic fertilizer (Yoshiba et al. 1998). Rice (polished) from organically farmed fields, certified by the Japan Agricultural Standards (JAS, Ministry of Agriculture, Forestry and Fisheries 2017a), from fields converted from conventional to organic farming following JAS regulations, and from conventional fields have been investigated; their mean δ^{15}N values were found to be +5.0‰, +3.5‰, and +2.8‰, respectively (Fujita et al. 2003). Thus, organic rice has a significantly higher δ^{15}N value than conventional rice. According to Fujita et al. (2003), although δ^{15}N values cannot provide a defined standard for identifying organic rice and conventional rice, these values indicate that rice with a δ^{15}N value lower than +3‰ is most likely grown conventionally (Fig. 1). A similar conclusion was reached by Suzuki et al. (2009) who found that the δ^{15}N values of organic rice (polished) collected across Japan ranged from +3.1 to +8.0‰ in organic rice, from +2.3 to +5.2‰ in rice grown under reduced synthetic fertilizer of less than half of that used in conventional cultivation, and from +0.3 to +3.2‰ in conventional rice. The δ^{15}N values of rice plants might quantitatively reflect the δ^{15}N values of N sources and the absorbed N from each source, namely soil, manure, and synthetic fertilizer (Tokunaga et al. 2000). This conclusion supports the idea that the δ^{15}N values of rice reflect N management regimes in the field.

δ^{15}N values of organic fertilizers

The δ^{15}N values of total N in 18 types of commercially available organic fertilizer in Akita Prefecture were determined (Nishida & Sato 2015). Finely ground samples were used for ^{15}N analysis. Two of the fertilizers were derived from plant processing waste, 11 from fish, livestock and plant processing wastes, four from chicken manure, and one from cattle manure. The selected fertilizers cover most types of organic fertilizer available on the market. Four synthetic fertilizers, ammonium sulfate, and three compound fertilizers that are commonly used in rice farming were also tested. The synthetic fertilizers were dissolved in distilled water. The solution was put into a tin capsule and freeze-dried, and then subjected to ^{15}N analysis.

Figure 2 shows the frequency distribution of the δ^{15}N values of organic and synthetic fertilizers. Also included are the δ^{15}N values of rice straw compost (+5.3‰, Nishida et al. 2007), livestock manure compost (+17.4‰, Nishida et al. 2007), and pelletized organic fertilizer made from

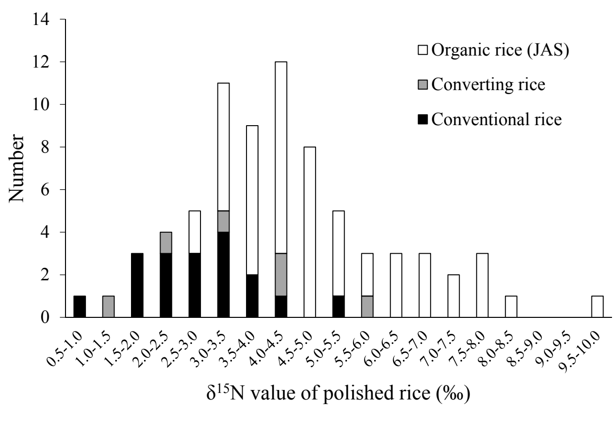

Fig. 1. Frequency distribution of natural ^{15}N abundance (δ^{15}N value) of rice (polished) collected from organic farming fields certified by Japan Agricultural Standards (JAS), fields converted from conventional to organic farming, and conventional fields in the Hokuriku region of Japan (Fujita et al. 2003)

n = 51, 6, and 18 for organic rice, converting rice, and conventional rice, respectively.

fish, livestock and plant processing wastes (+6.3‰) used at TARC/NARO. The δ^{15}N values of the organic fertilizers ranged from +2.3 to +17.4‰, with an average of +8.3‰. The δ^{15}N values of organic fertilizers made from livestock manure were relatively high ranging from +8.2 to +17.4‰, while those made from plant processing waste were relatively low ranging from +2.4 to +3.6‰, and those made from waste from fish, livestock and plant processing were intermediate, ranging from +4.3 to +12.9‰. Negative values were obtained for the synthetic fertilizers. The low δ^{15}N values of the synthetic fertilizers are consistent with the well-known fact that the δ^{15}N values of synthetic fertilizers are similar to atmospheric N, i.e., 0‰ (Yoneyama 1996). The reported δ^{15}N values for organic fertilizers are higher than those of synthetic fertilizers, ranging from +4.2‰ to +20.8‰ (Tokunaga et al. 2000, Nakano & Uehara 2006, Nishida et al. 2007, Nakano et al. 2010, Hayashi et al. 2011). Yoneyama et al. (1990) reported that alluvial soil, the dominant type of Japanese paddy soil, has a mean δ^{15}N value of +3.2‰ based on the δ^{15}N values of soils collected throughout Japan. Therefore, the δ^{15}N values of organic fertilizers generally tend to be higher than those of Japanese paddy soils, whereas those of synthetic fertilizers are likely to be lower.

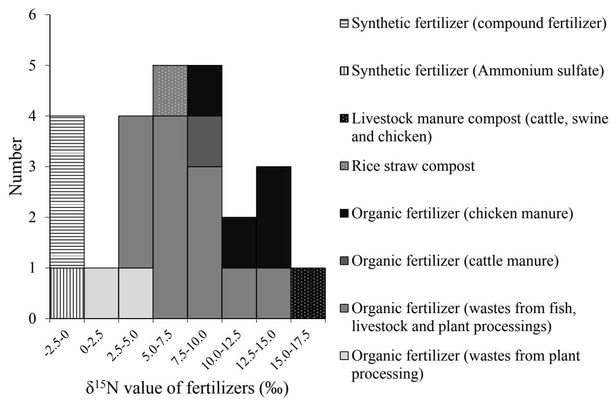

Fig. 2. Frequency distribution of natural ^{15}N abundance (δ^{15}N value) of organic fertilizers and synthetic fertilizers (Nishida & Sato 2015)

The δ^{15}N values of rice straw compost (+5.3‰), livestock manure compost (+17.4‰) and pelletized organic fertilizer (+6.3‰) used at the Tohoku Agricultural Research Center, National Agriculture and Food Research Organization located in Daisen, Akita, Japan are included. *n* = 4, 19, 1, and 1 for synthetic fertilizer, organic fertilizer, livestock manure compost, and rice straw compost, respectively.

Relationship between δ^{15}N values of rice and soil

Soil provides 60-70% of the N taken up by rice plants (Kyuma 2004). Consequently, soil may be the most influential factor on the δ^{15}N values of rice. Other N sources such as fertilizer can also influence the δ^{15}N values of rice; therefore, it is likely that organic farming will show a distinctive relationship between the δ^{15}N values of rice and soil.

1. δ^{15}N values of rice and soil in experimental fields at TARC/NARO

Nishida and Sato (2015) described the results of analyses conducted in experimental fields at TARC/NARO (N39°29′, E140°30′) located in Daisen, Akita, Japan from 2007 to 2009. The type of soil in the experimental fields is Gray Lowland Soil (Fluvisol, FAO 2006). The δ^{15}N values of total N in rice (unpolished) and soil were analyzed in a range of field conditions: application of rice straw compost, livestock manure compost, pelletized organic fertilizer, and/or synthetic fertilizer. Plots in which rice straw compost, livestock manure compost, or pelletized organic fertilizer was solely applied were considered to be organic in terms of fertilizer management. Finely ground samples were used for ^{15}N analyses in both rice and soil.

Regardless of the cropping year, the δ^{15}N values of rice tended to reflect those of the soil (Fig. 3). In all years, a significant positive correlation was observed between the δ^{15}N values of rice and soil in plots where no N source was applied. The δ^{15}N values of rice grown with only organic fertilizers were higher than the regression line for the δ^{15}N values of rice and soil without an applied N source. The δ^{15}N values of rice grown with synthetic fertilizers were lower than the regression line in 98% of the test plots screened in 2007-2009.

2. δ^{15}N values of rice and soil in commercially cultivated fields

Nishida and Sato (2015) also analyzed the δ^{15}N values of total N of rice (unpolished) and soil from commercially cultivated fields, both organically and conventionally farmed, in Daisen and Yokote (Akita Prefecture, Japan). The types of soil in Daisen are Gray Lowland Soil and Gley Lowland Soil (Fluvisol and Gleysol, FAO 2006), and those in Yokote are Gray Lowland Soil, Gley Lowland Soil, and Wet Andosol (Fluvisol and Gleysol, FAO 2006). For organic farming, only organic fertilizers were applied; for conventional farming, synthetic fertilizers were applied. Finely ground samples were used for ^{15}N analyses in both rice and soil.

The analyses gave the same results as obtained in the experimental fields, namely that the $\delta^{15}N$ values of rice reflected those of the soil (Fig. 4). Again, the $\delta^{15}N$ values of organic rice tended to be higher than the regression line for the $\delta^{15}N$ values of rice and soil obtained in the absence of an applied N source at TARC/NARO; the $\delta^{15}N$ values of conventional rice tended to be lower than the regression line. In 2007-2009, the $\delta^{15}N$ values of rice were higher than the regression line in 78-89% of organic rice samples; in contrast, those values were lower in 88-97% of conventional rice.

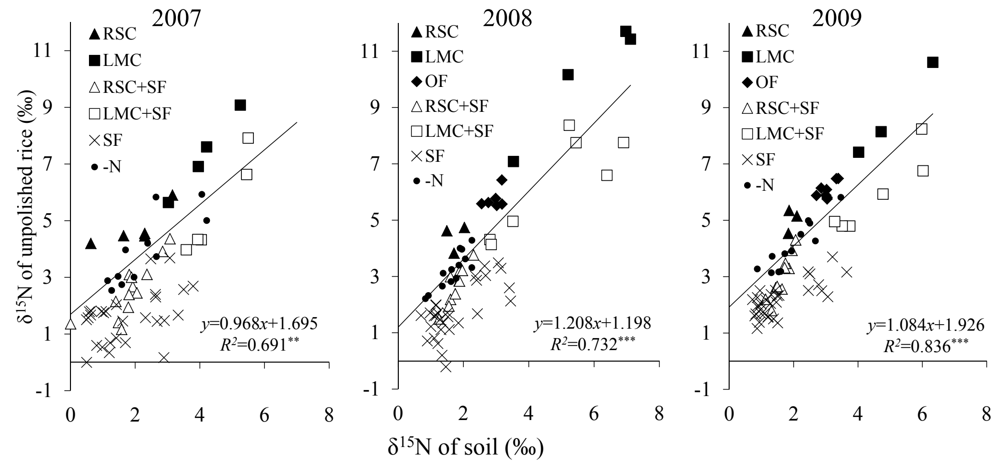

Fig. 3. Natural ^{15}N abundance ($\delta^{15}N$ value) of rice (unpolished) and soil in experimental fields at the Tohoku Agricultural Research Center, National Agriculture and Food Research Organization located in Daisen, Akita, Japan, and the regression line obtained from the $\delta^{15}N$ values of rice (unpolished) and soil without an N source application (–N) (Nishida & Sato 2015)

RSC: rice straw compost, LMC: livestock manure compost, OF: pelletized organic fertilizer, RSC+SF: rice straw compost and synthetic fertilizer, LMC+SF: livestock manure compost and synthetic fertilizer, SF: synthetic fertilizer, –N: no nitrogen source. $^{**}P < 0.01$, $^{***}P < 0.001$. n = 5, 4, 13, 5, 28, and 11 in RSC, LMC, RSC+SF, LMC+SF, SF, and -N, respectively, in 2007; n = 3, 4, 6, 10, 7, 29, and 13 in RSC, LMC, OF, RSC+SF, LMC+SF, SF, and -N, respectively, in 2008; n = 3, 3, 8, 10, 6, 32, and 12 in RSC, LMC, OF, RSC+SF, LMC+SF, SF, and -N, respectively, in 2009.

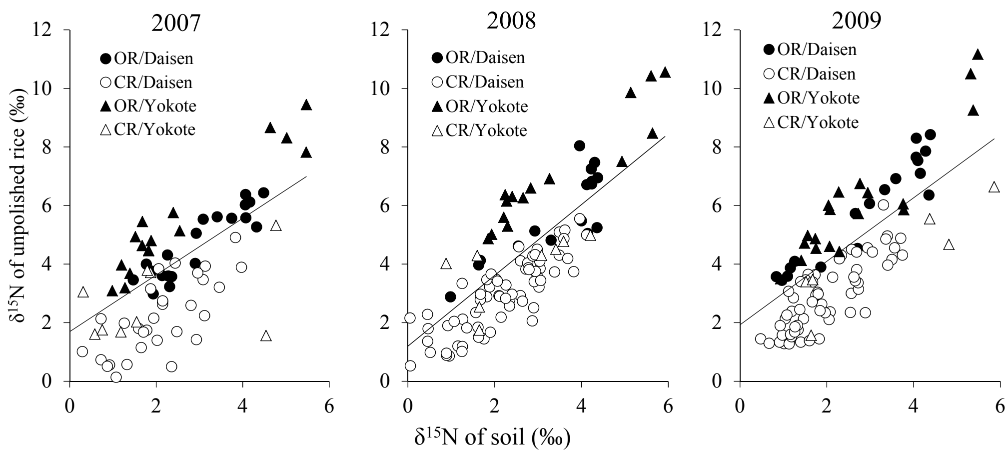

Fig. 4. Natural ^{15}N abundance ($\delta^{15}N$ value) of organic rice (unpolished), conventional rice (unpolished) and soil in commercial fields in Daisen and Yokote, Akita, Japan, and the regression line obtained from the $\delta^{15}N$ values of rice (unpolished) and soil without an N source application in experimental fields at the Tohoku Agricultural Research Center, National Agriculture and Food Research Organization located in Daisen, Akita, Japan (Nishida & Sato 2015)

OR: organic rice, CR: conventional rice. n = 20, 31, 16, and 9 in OR/Daisen, CR/Daisen, OR/Yokote, and CR/Yokote, respectively, in 2007; n = 16, 67, 15, and 8 in OR/Daisen, CR/Daisen, OR/Yokote, and CR/Yokote, respectively, in 2008; n = 18, 62, 19, and 6 in OR/Daisen, CR/Daisen, OR/Yokote, and CR/Yokote, respectively, in 2009.

3. Discrimination of organic and conventional rice using relationship between δ15N values of rice and soil

The analyses described above indicate that it may be possible to use the relationship in the δ15N values of rice and soil without an applied N source, in order to distinguish between organic and conventional rice. This approach is effective if the relative δ15N values descend in the order of organic fertilizer, soil, and synthetic fertilizer. The δ15N values of organic fertilizers used at TARC/NARO were higher than the δ15N values of soils in the experimental plots (Figs. 2 and 3). Similarly, the δ15N values of most commercial organic fertilizers are higher than the δ15N values of soils in the fields at Daisen and Yokote (Figs. 2 and 4). Hence, the δ15N values of rice grown with these organic fertilizers would be expected to be higher than those of rice grown without an N source application. In contrast, the δ15N values of the synthetic fertilizers were negative (Fig. 2); therefore, rice grown with these fertilizers would be expected to have lower δ15N values than those of rice grown without an N source application. If both organic and synthetic fertilizers were applied, the δ15N values of rice would vary according to the relative rates of application of both types of fertilizer, their δ15N values, and their N efficiencies for rice plants. The N efficiencies of synthetic fertilizers are generally higher than those of organic fertilizers (Nishida et al. 2004, Chalk et al. 2013). Consequently, the influence of a synthetic fertilizer N with a low δ15N value is likely to be more apparent than those of organic fertilizers. Figure 5 shows the relationship between the δ15N values of unpolished rice and those of polished rice collected at TARC/NARO in 2006, 2007, and 2008. The δ15N values of unpolished rice and polished rice are almost identical. Thus, both unpolished rice and polished rice can be applied to the discrimination approach to distinguish between organic rice and conventional rice using the relationship between the δ15N values of rice and those of soil.

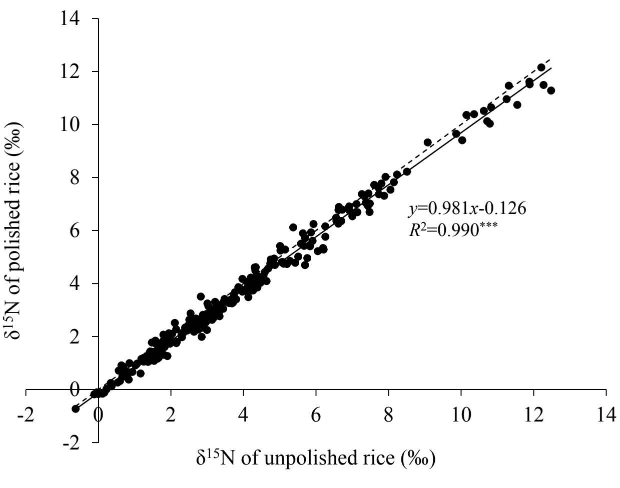

Fig. 5. Relationship between natural 15N abundance (δ15N value) of unpolished rice and those of polished rice collected at the Tohoku Agricultural Research Center, National Agriculture and Food Research Organization located in Daisen, Akita, Japan in 2006, 2007 and 2008

The solid line indicates the regression line between the δ15N values of unpolished rice and those of polished rice. n = 254. ***P < 0.001. The dashed line indicates y = x.

The available information suggests that it would be difficult to use a particular δ15N value for rice as an indicator of organic growth conditions. Fujita et al. (2003) proposed a threshold value of +3‰, suggesting that rice with a value < +3‰ was most likely produced by conventional cultivation. However, as can be seen in Figs. 3 and 4, conventionally grown rice samples can have δ15N values higher than +3‰. The discriminant approach based on the relationship of the δ15N values of rice and soil is feasible for any range of δ15N values in rice.

Regional variation in the relationship between δ15N values of rice and soil

1. Existence of regional variation in the relationship between δ15N values of rice and soil

In order to apply the discriminant approach based on the δ15N values of rice and soil in other areas, it is essential to determine the existence (or otherwise) of regional variation in the relationship. Nishida and Sato (2016) undertook a study to investigate possible regional variation by comparing the relationship between the δ15N values of rice and unamended soil at Ogata Farm, Akita Agricultural Experiment Station (N40°01′, E139°58′) with those at TARC/NARO (Nishida & Sato 2015). The two sites are approximately 70 km apart. A significant positive correlation was observed between the δ15N values of rice and soil in Ogata (Fig. 6), but differed from that observed in Daisen. An analysis of covariance (ANCOVA) showed that the regression slopes did not differ significantly, but a significant effect of the site on the δ15N values of rice was evident when the common slope (1.01) was applied. This indicates that the δ15N values of rice in Daisen are significantly higher than those in Ogata. The difference between the intercepts of the regression lines was 0.8‰. These results showed the existence of regional variation in the relationship between the δ15N values of rice and those of unamended soil.

2. Possible reasons for regional variation

The δ[15]N values of the natural N input may be a factor that affects the relationship between the δ[15]N values of rice and those of unamended soil. When no N fertilizer is applied, the N sources for rice plants are soil, irrigation water, wet deposit (rain), and biological N fixation. Nitrogen from irrigation water may be one of the reasons for the higher δ[15]N values of rice in Daisen than in Ogata. In 2009, the δ[15]N values of irrigation water at Daisen during the growing season ranged from +3.7 to +4.9‰ and averaged +4.3‰. In contrast, the values at Ogata were significantly lower, ranging from +1.1 to +2.1‰ and averaging +1.5‰. Taking the level of higher precipitation in Daisen and the negative δ[15]N values of rain (Hayasaka et al. 2005, Wo et al. 2008) into account, N from rainfall is not considered a factor in the higher δ[15]N values of rice observed at Daisen compared to those at Ogata. Biological N fixation is also not considered a factor because a greater amount of biological N fixation (Yasuda et al. 2000) with negative δ[15]N values (Yoneyama 1987) can be expected in Daisen.

Another possible factor that affects the δ[15]N values of mineralized N in soil is ammonia nitrification in the oxidized layer, followed by denitrification of nitrates in the reduced layer. Both processes can enrich the [15]N content of the remaining mineralized N in the soil. Nitrification-denitrification processes may occur over a longer period in Daisen, which has higher water permeability than Ogata (Hasebe et al. 1986), resulting in the enrichment of [15]N in rice in Daisen. This process may therefore be a factor in the higher observed δ[15]N values of rice in Daisen than in Ogata. Thus, the difference in the δ[15]N value of natural N inputs and N transformation in the soil in the target area should be carefully considered when employing the relationship between the δ[15]N values of rice and those of soil to discriminate between organic and conventional rice.

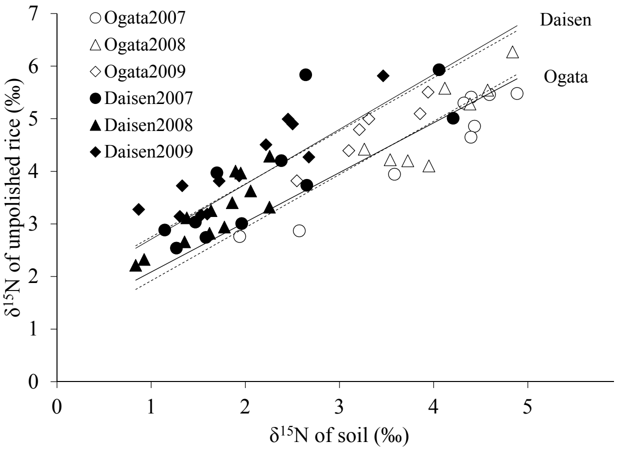

Fig. 6. Relationship between natural [15]N abundance (δ[15]N value) of rice (unpolished) and those of soil without an applied N source in Ogata and Daisen, Akita, Japan (Nishida & Sato 2016)

The solid lines indicate the regression lines for the original data (Ogata, $y = 0.95x + 1.15$, $R^2 = 0.716$***, n=23; Daisen, $y = 1.04x + 1.66$, $R^2 = 0.711$***, n = 36). ***$P < 0.001$. The dashed lines indicate the regression lines applied using the common slope based on ANCOVA (Ogata, $y = 1.01x + 0.91$; Daisen, $y = 1.01x + 1.74$).

Temporal variation in relationship between δ[15]N of rice and soil

In addition to regional variation, temporal variation in the relationship between the δ[15]N values of rice and those of soil has been tested. At each site, ANCOVA showed that the regression slopes did not differ significantly due to a lack of significant interaction, but a significant effect of the cropping year on the δ[15]N values of rice was evident when applying the common slope (1.03 in Daisen, 1.09 in Ogata) (Table 1). These results showed the existence of temporal variation in the relationship between the δ[15]N values of rice and those of unamended soil. The cause of such temporal variation is

Table 1. Analysis of covariance (ANCOVA) results for testing the effect of cropping year on the relationship between natural [15]N abundance (δ[15]N value) of rice and soil without N source application

		Include interaction			Common slope applied		
		df	*F*	*P*	df	*F*	*P*
Daisen	Cropping year	2	3.39	0.047	2	4.52	0.019
	δ[15]N value of soil	1	65.28	< 0.0001	1	88.79	< 0.0001
	Cropping year x δ[15]N value of soil	2	0.28	0.755			
Ogata	Cropping year	2	6.52	0.008	2	10.33	0.001
	δ[15]N value of soil	1	67.63	< 0.0001	1	114.11	< 0.0001
	Cropping year x δ[15]N value of soil	2	0.45	0.643			

n = 11, 13, and 12 in 2007, 2008, and 2009, respectively, in Daisen. n = 9, 8 and 6, in 2007, 2008, and 2009, respectively, in Ogata.

unknown at present. However, the results suggest that the regression line between the δ15N values of rice and those of unamended soil need to be identified in each cropping season, in order to use the relationship between the δ15N values of rice and soil to discriminate between organic and conventional rice.

Perspectives

As described in this review, the relationship between the δ15N values of rice and soil can be used to discriminate between organic rice and conventional rice. This approach may aid in enhancing both food safety and security. However, δ15N values alone cannot completely discriminate between organic rice and conventional rice. Therefore, other approaches need to be developed. A combination of multiple methods (e.g., trace element component) will enable a more sensitive means of identifying organic and conventional rice.

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