Characteristics of the Relationship between Natural $^{15}\text{N}$ Abundances in Organic Rice and Soil

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Abstract: The characteristics of the relationship of natural $^{15}\text{N}$ abundances ($\delta^{15}\text{N}$ 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 using this relationship was explored. Organically grown rice, conventionally grown rice and soil samples were collected from farmers’ fields in Daisen city and Yokote city, Akita prefecture, Japan as well as from the experimental fields of NARO Tohoku Agricultural Research Center located in Daisen from 2007 to 2009. Organic fertilizers and synthetic fertilizers available on the market in Akita prefecture were also collected. The $\delta^{15}\text{N}$ values of those samples were measured. Regardless of the farming method, the $\delta^{15}\text{N}$ values of rice reflected those of the soil. The $\delta^{15}\text{N}$ values of the organic fertilizers available on the market were likely to be higher than those of the soil. Meanwhile the $\delta^{15}\text{N}$ values of the synthetic fertilizers were negative and lower than those of the soil. The $\delta^{15}\text{N}$ values of organically grown rice tended to be higher than the regression line obtained from the $\delta^{15}\text{N}$ values of rice and soil without N source application. The $\delta^{15}\text{N}$ values of conventionally grown rice tended to be lower than the regression line. These results indicated that the relationship of the $\delta^{15}\text{N}$ values of rice and soil without an applied N source could aid in discriminating between organic rice and conventional rice.

Key words: Natural $^{15}\text{N}$ abundance, Organic farming, Organic fertilizer, Paddy soil, Rice, Synthetic fertilizer.

Demand for organic food, including rice ($\text{Oryza sativa}$ L.), is growing. Certified organic rice production in Japan increased from 7,777 Mg in 2001 to 10,342 Mg in 2012 (Ministry of Agriculture, Forestry and Fisheries, 2014a). Meanwhile, mislabeled organic rice, which was grown conventionally using synthetic fertilizer, has been traded as organic rice in the marketplace (Ministry of Agriculture, Forestry and Fisheries, 2008). Recently, food frauds appear in various types of food including mislabeled organic rice and have become a major social problem (Ministry of Agriculture, Forestry and Fisheries, 2014b). Because the food frauds endanger the safety and security of food, the development of preventive measures against the food frauds is an important issue. An objective and scientific method to discriminate between organic and conventional (nonorganic) rice needs to be developed to prevent cases of misrepresentation.

Methods for residue analysis of pesticides have been developed (Tanizawa et al., 2005; Kobayashi, 2009). Regarding nutrient management regime, natural $^{15}\text{N}$ abundance ($\delta^{15}\text{N}$ value) has received attention as an indicator to discriminate organically grown crops from conventionally grown crops. The $\delta^{15}\text{N}$ values of organically grown crops without synthetic fertilizer tend to be higher than those of conventionally grown crops with synthetic fertilizer, as observed in some vegetables (Nakano et al., 2002, 2003b, 2010; Bateman et al., 2005, 2007; Nakano and Uehara, 2005, 2006a; Yun et al., 2006; Camin et al., 2007, 2011; Amor et al., 2008; Rogers, 2008; Šturm and Lojen, 2011; Šturm et al., 2011), fruits (Camin et al., 2011), and tea (Morita et al., 1999; Hayashi et al., 2011). $\delta^{15}\text{N}$ values have also been reported to be higher in organic rice than in conventional rice (Fujita et al., 2003; Suzuki et al., 2009), and a threshold value of $3\%_{\text{o}}$ has been proposed (Fujita et al., 2003). However, the validity of this threshold value has not been fully assessed. Furthermore, the relationship of $\delta^{15}\text{N}$ values between rice grown organically or conventionally and their respective soils has not been investigated.

Soil is the major nitrogen (N) source for rice plants which accounts for 60 to 70% of total N taken up by the...
rice plant (Shoji and Mae, 1984). Then the soil may be one of the influential factors for the $\delta^{15}$N value of rice, suggesting the importance of the relationship of the $\delta^{15}$N values between rice and soil. Other N sources, such as organic materials and synthetic fertilizer, may also affect the $\delta^{15}$N value of rice. Organic farming might leave a distinctive signature in the relationship of $\delta^{15}$N values between rice and soil. This study was performed to investigate characteristics of the relationship of $\delta^{15}$N values between rice and soil in organic and conventional farming, and to explore the possibility of discriminating organic rice from conventional rice using the relationship.

Materials and methods

1. Sample collection

Rice and soil samples were collected from the experimental fields in NARO Tohoku Agricultural Research Center (NARO/TARC, 39°29′ N, 140°30′ E) located in Daisen city, Akita prefecture, Japan in 2007, 2008 and 2009. In the same years, rice and soil samples were also collected from farmers’ fields where organic or conventional farming was conducted in Daisen city (39°27′ N, 140°29′ E at the city office) and Yokote city (39°19′ N, 140°34′ E at the city office), Akita prefecture, northeast Japan. In organic farming, rice plants were grown with application of only organic fertilizers without synthetic fertilizer. In conventional farming, rice plants were grown with application of synthetic fertilizer. The soil type of the paddy fields in the study area is alluvial soil [Fluvisol and Greysol (FAO, 2006)].

In the fields of NARO/TARC, experiments on the application of rice straw compost, livestock manure compost, organic fertilizer and synthetic fertilizer were performed for multiple studies as listed in Table 1. The rice straw compost was produced at NARO/TARC by piling with occasional turnover in a composting house. In the plot where only organic materials, namely rice straw compost, livestock manure compost or pelletized organic fertilizer, was applied, rice plants were organically grown in terms of fertilizer management. Then in the present study, rice from these plots was regarded as organically grown rice. In each plot, rice plants were harvested from the area of approximately 3.3 m$^2$ at the maturity stage. Topsoil was collected by an auger consisting of a stainless steel tube 3 cm in diameter around the harvesting location. The topsoil thickness was approximately 15 cm. The varieties of rice used were Akitakomachi. In some plots, samples were collected every year (three times), and in other plots, sample collection was performed only once or twice.

In the farmers’ fields in Daisen and Yokote, four hills of rice plants (about 0.2 m$^2$) were harvested from two

| Year | Daisen | Conventional farming | Yokote | Conventional farming |
|------|--------|----------------------|--------|----------------------|
| 2007 | 20     | 31                   | 16     | 9                    |
| 2008 | 16     | 67                   | 15     | 8                    |
| 2009 | 18     | 62                   | 19     | 6                    |

RSC, only rice straw compost was applied; LMC, only livestock manure compost was applied; OF, only pelletized organic fertilizer was applied; RSC + SF, both rice straw compost and synthetic fertilizer were applied; LMC + SF, both livestock manure compost and synthetic fertilizer were applied; SF, only synthetic fertilizer was applied; –N, no nitrogen source was applied.

| Treatment | Year | RSC | LMC | OF | RSC + SF | LMC + SF | SF | –N |
|-----------|------|-----|-----|----|----------|----------|----|----|
| 2007      |      | 5   | 4   | 0  | 13       | 5        | 28 | 11 |
| 2008      | 3    | 4   | 6   | 10 | 7        | 29       | 13 |
| 2009      | 3    | 3   | 8   | 10 | 6        | 32       | 12 |
These synthetic fertilizers were common types in rice fertilizer. They contained 15% of N, P\(\text{2O}_5\) and K\(\text{2O}\), respectively. Another compound pentoxide equivalent, P\(\text{2O}_5\)) and potassium (in potassium fertilizer) were also collected. Two compound synthetic fertilizer, namely, ammonium sulfate and three available on the market in the study area. Four types of and one made from cattle manure. These organic fertilizers were collected in 2008. Two types were made from wastes from fish, livestock plant processing, four types from chicken manure, plant processing, 12 types from wastes from fish, livestock and one made from cattle manure. These organic fertilizers could cover most types of organic fertilizer available on the market in the study area. Four types of synthetic fertilizer, namely, ammonium sulfate and three compound fertilizers were also collected. Two compound fertilizers contained 13% of N, phosphorus (in phosphorus pentoxide equivalent, P\(\text{2O}_5\)) and potassium (in potassium oxide equivalent, K\(\text{2O}\)), respectively. Another compound fertilizer contained 15% of N, P\(\text{2O}_5\) and K\(\text{2O}\), respectively. These synthetic fertilizers were common types in rice farming.

Separate areas in each field at maturity stage. Around the harvesting location, topsoil was collected by the auger. The topsoil thickness was around 10 – 15 cm. Samples from each place were individually analyzed and averaged for a representative value of each field. In Daisen, the number of organic farming fields where samples were collected was 20, 16, and 18 in 2007, 2008 and 2009, respectively, and the number of conventional farming fields was 31, 67, and 62 in 2007, 2008 and 2009, respectively (Table 2). In Yokote, the number of organic farming fields where samples were collected was 16, 15 and 19 in 2007, 2008 and 2009, respectively, and the number of conventional farming fields was nine, eight and six in 2007, 2008 and 2009, respectively (Table 2). The rice variety was Akitakomachi. In some fields, sample collection was conducted every year (three times), and in other fields, sample collection was performed only once or twice.

Nineteen types of commercial organic fertilizer were collected in 2008. Two types were made from wastes from plant processing, 12 types from wastes from fish, livestock and plant processing, four types from chicken manure, and one made from cattle manure. These organic fertilizers could cover most types of organic fertilizer available on the market in the study area. Four types of synthetic fertilizer, namely, ammonium sulfate and three compound fertilizers were also collected. Two compound fertilizers contained 13% of N, phosphorus (in phosphorus pentoxide equivalent, P\(\text{2O}_5\)) and potassium (in potassium oxide equivalent, K\(\text{2O}\)), respectively. Another compound fertilizer contained 15% of N, P\(\text{2O}_5\) and K\(\text{2O}\), respectively. These synthetic fertilizers were common types in rice farming.

Fig. 1. Natural \(\delta^{15}N\) abundance (\(\delta^{15}N\) value) of rice (\textit{Oryza sativa L.}) and soil in experimental fields at NARO Tohoku Agricultural Research Center located in Daisen, Akita, Japan, and the regression line obtained from the \(\delta^{15}N\) values of rice and soil without an N source application (–N). RSC, only rice straw compost was applied; LMC, only livestock manure compost was applied; SF, only peletized organic fertilizer was applied; RSC+SF, both rice straw compost and synthetic fertilizer were applied; LMC+SF, both livestock manure compost and synthetic fertilizer were applied; SF, only synthetic fertilizer was applied; –N, no nitrogen source (neither organic material nor synthetic fertilizer) was applied. **P<0.01, ***P<0.001.

2. Sample preparation and analysis

Rice plants were threshed and hulled. The brown rice (hulled rice) was finely ground with a vibrating mill (TI-100, C.M.T., Saitama, Japan). The soil was air-dried at room temperature, passed through a 2-mm sieve and finely ground with the vibrating mill. Organic fertilizers were oven-dried at 70°C, then finely ground with the vibrating mill. These samples were put into tin capsules and analyzed for natural \(\delta^{15}N\) abundance using an isotope-ratio mass spectrometer (IRMS) coupled with an elemental analyzer (DeltaXP, Thermo Fisher Scientific Inc., Waltham, MA, USA). The synthetic fertilizers were dissolved in distilled water. The solution was put into a tin capsule and freeze-dried. They were then analyzed for natural \(\delta^{15}N\) abundance using the IRMS coupled with an elemental analyzer.

Nitrogen isotope data are reported in conventional \(\delta\) notation in units of per mil (‰) with respect to atmospheric N according to the following equation:

\[\delta^{15}N_{\text{sample}}(\text{‰}) = \left(\frac{R_{\text{sample}}}{R_{\text{air}}} - 1\right) \times 1000,\]

where \(R_{\text{sample}}\) and \(R_{\text{air}}\) are \(^{15}N/^{14}N\) ratios of the measurement sample and the air, respectively.

3. Statistical analysis

Regression analysis was conducted between \(\delta^{15}N\) values of rice and soil in the plots where no N source (neither organic material nor synthetic fertilizer) was applied in NARO/TARC using JMP 9.0 (SAS Institute, 2010).

Results

The \(\delta^{15}N\) values of rice and soil at NARO/TARC are shown in Fig. 1. The \(\delta^{15}N\) value of soil at NARO/TARC
ranged from 0 to 5.5‰, 0.7 to 7.1‰ and 0.7 to 6.3‰ in 2007, 2008 and 2009, respectively. The δ¹⁵N value of rice at NARO/TARC ranged from 0 to 9.1‰, –0.2 to 11.7‰ and 1.2 to 10.6‰ in 2007, 2008 and 2009, respectively. Regardless of the cropping year, the δ¹⁵N values of rice tended to be higher than the regression line. The δ¹⁵N values of rice were 2.3, 3.1 and 3.0‰ in 2007, 2008 and 2009, respectively.

In the farmers’ fields as well, the δ¹⁵N values of rice reflected those of soil (Fig. 2). The δ¹⁵N values of organic rice tended to be higher than the regression line obtained from the δ¹⁵N values of rice and soil without an applied N source at NARO/TARC; and the δ¹⁵N values of conventional rice tended to be lower than the regression line. The δ¹⁵N values of 28 out of 36 (78%), 27 out of 31 (87%), and 33 out of 37 (89%) organic rice grown in 2007, 2008 and 2009, respectively, were higher than the regression line. The δ¹⁵N values of 36 out of 40 (90%), 66 out of 75 (88%), and 66 out of 68 (97%) conventional rice grown in 2007, 2008 and 2009, respectively, were lower than the regression line.

The frequency distribution of the δ¹⁵N values of organic fertilizers and synthetic fertilizers is shown in Fig. 3. The results include the δ¹⁵N values of rice straw compost (5.3‰, Nishida et al., 2007) and livestock manure compost (17.4‰, Nishida et al., 2007) used at NARO/TARC. The δ¹⁵N values of organic fertilizers including rice straw and livestock manure composts ranged from 2.3 to 17.4‰ with an average of 8.3‰. The δ¹⁵N values of organic fertilizers made from livestock manure were relatively low. The δ¹⁵N values of organic fertilizers made from wastes from plant processing were relatively high, and those of organic fertilizers made from wastes from plant processing were relatively low. The δ¹⁵N values of organic fertilizers made from wastes from fish, livestock and plant processings were likely to be intermediate. The δ¹⁵N value of pelletized organic fertilizer used at NARO/TARC was 6.3‰. The δ¹⁵N values of the synthetic fertilizers were negative.

**Discussion**

The results of the present study suggest that the relationship of the δ¹⁵N values of rice and soil without an applied N source can aid in discriminating between
organic fertilizers available on the market were higher than that of soil and synthetic fertilizer. In many cases, the fertilizer was applied (Figs. 1 and 3). The fertilizer values for rice grown with these organic fertilizers could be higher than those of rice without an N source application. In contrast, the $\delta^{15}N$ values of rice grown with a low application rate of organic fertilizers were lower than those of rice without an applied N source. Hence, the $\delta^{15}N$ values of rice grown with synthetic fertilizers is generally higher than those of organic fertilizers. The N efficiency of synthetic fertilizers has been reported to be as low as atmospheric N (Shearer et al., 1973; Freyer and Ali, 1974; Black and Waring, 1977; Yoneyama, 1996; Bateman and Kelly, 2007). Rice grown with the synthetic fertilizer would be lower than those of the rice grown without an N source application. In the plots where both organic fertilizers and synthetic fertilizers were applied, the $\delta^{15}N$ values of rice might vary with the application rates of organic fertilizers and synthetic fertilizers, their $\delta^{15}N$ values, and their N efficiencies for rice plants. The N efficiency of synthetic fertilizers is generally higher than those of organic fertilizers (Nishida et al., 2004; Chalk et al., 2013). Consequently, the influence of the synthetic fertilizer N with a low $\delta^{15}N$ value was likely to be more apparent than those of organic fertilizers.

This discriminant approach is effective when the relative descending order of the $\delta^{15}N$ values of organic fertilizer, soil and synthetic fertilizer. In many cases, the $\delta^{15}N$ values of organic fertilizers are likely to be higher than those of the Japanese paddy soil; and the $\delta^{15}N$ values of synthetic fertilizers would be lower than those of the Japanese paddy soil. A previous study showed that the mean $\delta^{15}N$ value of the alluvial soil in Japan was 3.2‰ (Yoneyama et al., 1990), which was the dominant type of Japanese paddy soil (Wada, 1984). $\delta^{15}N$ values of synthetic fertilizers have been reported to be as low as atmospheric N (Shearer et al., 1973; Freyer and Ali, 1974; Black and Waring, 1977; Yoneyama, 1996; Bateman and Kelly, 2007). On the other hand, the reported $\delta^{15}N$ values for organic fertilizers in Japan were high, ranging from 4.2‰ to 20.8‰ (Morita et al., 1999; Tokunaga et al., 2000; Nakano et al., 2003a, 2003b, 2010; Nakano and Uehara, 2005, 2006b, 2009; Nishida et al., 2007; Hayashi et al., 2011).

The regression lines of the $\delta^{15}N$ values of rice and soil without an applied N source were similar for 3 years (Fig. 1). However, these lines were not fully identical and this implied that the relationship between the $\delta^{15}N$ values of rice and soil could vary somewhat with cropping year.

The results of the present study suggest difficulty in using a particular $\delta^{15}N$ value for rice as an indicator of organic growth conditions. Fujita et al. (2003) proposed a threshold value of 3‰, which meant rice with a $\delta^{15}N$ value lower than 3‰ is most likely conventionally grown rice. The results of the present study indicated that this threshold value was incomplete, because there were many conventionally grown rice samples with $\delta^{15}N$ values higher than 3‰.

Further study is needed to assess the validity of this discriminant approach in other areas and to clarify the
cause for annual variation of the relationship between the $\delta^{15}$N values of rice and soil. Additionally, other methods to help discriminate organic rice from conventional rice are expected to be developed. A combination of multiple methods will enable a more sensitive discrimination between organic and conventional rice.

Acknowledgements

We thank Dr. Kondo M. for $^{15}$N analysis, Mr. Sato M., Mr. Toyokawa S. and farmers for their cooperation with sample collection, Dr. Yoshida K. and Dr. Sekiya H. for helpful suggestions.

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* In Japanese with English abstract.
** In Japanese.