REVIEW

Agricultural Product Authenticity and Geographical Origin Traceability

- Use of Nondestructive Measurement -

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Abstract

As the security of agricultural products has become a serious issue faced by people worldwide in recent decades, measures for an effective supervision system for agricultural product quality and safety are continuously being developed. In such a system, the assessment of agricultural product authenticity and geographical origin traceability could play a very important role. Recently, researchers have been focusing on some successful techniques, including the stable isotopic technique, compositional analysis technique, spectroscopic technique, and sensor technology. The benefits from advances made in spectroscopy and imaging technology have facilitated the development of imaging spectrometry techniques that offer such advantages as being nondestructive, rapid, and requiring minimal to no sample preparation. This paper discusses several nondestructive technologies used for the assessment of agricultural product authenticity and geographical origin traceability, with a special focus on the nondestructive technology of imaging using an electromagnetic spectrum for agricultural product safety and quality. It specifically discusses the technology of ultraviolet imaging, hyperspectral imaging, fluorescence spectrometry, nuclear magnetic resonance, and terahertz spectroscopy according to different wavelengths and frequency of this electromagnetic spectrum. Although the application of nondestructive measurements performed using the electromagnetic spectrum for identifying agricultural product authenticity and geographical origin traceability is increasing along with advanced technology and lower equipment costs, the accuracy of such measurements must still be improved and the advantages in practical applications need to be determined.

Discipline: Agricultural engineering **Additional key words:** electromagnetic spectrum, infrared, terahertz, ultraviolet, visible

Introduction

In recent decades, food security has become a serious issue for people in developed as well as developing countries; thus, systems for agricultural production, processing, storage, and transportation are being developed to ensure safe food supply. On one hand, with the development of technology and growing attention on human health, the standards of food safety and quality are continuously being improved with the introduction of new food safety and quality supervision rules; on the other hand, with the development of technology, "food adulteration" is diversely increasing over time. Therefore, new analytical techniques to identify food or food ingredients are urgently needed. These demands have thus promoted the

*Corresponding author: e-mail anakano@affrc.go.jp Received 4 April 2017; accepted 31 August 2017. development of food safety and quality measurement techniques. And considerably more consideration is given to the quality and safety of agricultural products, as the main part of food supply, especially with regard to their authenticity and geographical origin traceability.

The quality and safety measurement of agricultural products are divided into two stages that include traditional manual measurement (e.g., classical methods, wet chemical methods) and instrumental analysis (Table 1). Along with technological advances, almost all manual measurements were substituted by instrumental analysis, such as electrochemical analysis (Bard & Faulkner 2000, Skoog et al. 1988), spectroscopy technology (Zou X. & Zhao J. 2015), chromatography technology (Zparg Z. et al. 2016), mass spectrometry technology (Sparkman 2000, Downard 2004,

Methods	Main application in food authenticity and traceability			
Stable isotope ratio mass spectrometry	Food authentication and traceability			
DNA methods	Identifying plant and animal species in food			
Polymerase chain reaction technique	Identifying food components			
Enzyme immunoassays	Identifying animal species in food			
Enzymatic techniques	Authenticating food components			
Proteome and metabolite	Analyses for food authentication			
Chemometrics	Data analysis			
Gas chromatography	Food analysis			
High-pressure liquid chromatography	Food authentication			
In-line sensors	Food analysis			
Near infra-red absorption technology	Analyzing food composition			
Nuclear magnetic resonance spectroscopy	Food authentication			
Spectrophotometric techniques	Food authentication and traceability			

Table 1. Common methods used for agricultural authenticity and traceability

Kondo N. et al. 2016), biochip technology (Fan et al. 2009), and stable isotope analysis (Yuan Y. et al. 2014, Nakano A. & Uehara A. 2008, 2009; Nakano A. et al. 2010, Nakano & Zhao 2018). However, several factors limit the efficiency of instrumental analysis for agricultural product assessment. For example, pretreatment-the key process in agricultural product quality and safety measurement-is necessary in both classical methods and instrumental analysis. Pretreatment usually cannot satisfy the demand for rapid, real-time, and field-level measurements in practical applications due to its complex and time-consuming process, and the high requirements for instruments. Even for the most advanced instrumental analysis methods, such issues as a long measurement period, high cost, and the professional expertise required still exist. Researchers have been developing new and more efficient approaches that can be applied over a wide range, and which offer sufficiently high accuracy to analyze known or unknown product components and element composition, and even microbial or chemical contaminants. Recent advances in electronics have facilitated the design of considerably more convenient probes and instruments with high resolution, which have diversified the applications of nondestructive measurements in food science and technology, thereby enhancing food quality and safety. Nondestructive measurements are now being recognized and adopted in agricultural product testing.

Nondestructive measurement

Nondestructive measurement technology encompasses a wide group of analysis techniques used to evaluate the quality and safety of food or agricultural products without causing damage (Cartz 1995), and is a very valuable technology that is both cost- and time-effective in product

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evaluation, processing, and research. Fundamental nondestructive measurement technology uses the changing relation between input energy (excitation) and output energy (feedback) to detect internal and external information regarding an object (Deng & Liu 2011). First, when external energy is applied to an object, the optical, force, acoustic and electrical energies will be influenced differently according to the object's properties, leading to different energies being output by an associate sensor. Next, a mathematic model for object qualitative or quantitative analysis is developed using the external energy information obtained and the physical or chemical information regarding the object's quality characteristics. Nondestructive measurement technology provides information about product properties such as structure, dimensions, and metrology, physical and mechanical properties, composition and chemical analysis, stress and dynamic responses, and signature analysis (Zou & Zhao 2015).

With the development of spectroscopy and imaging technologies, the application of imaging within the electromagnetic spectrum in agricultural product safety and quality has been increasing annually (Table 2); however, it is still less applied in agricultural product authenticity and geographical origin traceability. This study focuses on the application of imaging in agriculture production systems by using different regions of the electromagnetic spectrum, including ultraviolet, visible, near-infrared, mid-infrared, and terahertz. Table 2 and Figure 1 show the results obtained over a very wide frequency range and wavelength $(10^{-12} \text{ m to } 10^6 \text{ m})$.

1. Gamma-ray and X-ray imaging

Both gamma rays (wavelength: <0.01 nm) and X-rays (wavelength: 0.01-10 nm) have high frequency and energy,

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Electromagnetic spectrum imaging techniques	Natural wavelength or range of excitation	Sensors or devices	pplication in agricultural products		
Gamma Ray	0.01 nm	Scintillation count	Food irradiation Promote mutation Decrease infestation		
X-Ray Imaging	0.01–10 nm	Amorphous silicon	Monitor food quality Monitor composition Food irradiation		
Fluorescence	RF excitation	Fluorometers	Food geographic Monitor food safety Monitor composition		
Visual Imaging	400–700 nm	CCD or CMOS Camera	Monitor plant stress Monitor food quality Monitor food safety		
Multi-spectral Imaging	400–2500 nm	CCD & IR Camera	Food analysis Detect plant stress Monitor food safety		
NIR Spectroscopy	800–2500 nm	Spectrometer	Food analysis Detect plant stress Monitor food safety		
Thermal Imaging	8000–15000 nm	Thermal camera	Detect water stress Monitor food quality		
Terahertz	10^{5} - 10^{6} nm	Terahertz spectroscopy	Monitor plant stress Monitor food safety		
NMR Imaging	RF excitation	NMR scanner	Monitor toxins in food Monitor food quality Monitor food safety		

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RF: Radio Frequency, CCD: Charge-Coupled Device, COMS: Complementary Metal-Oxide-Semiconductor transistor, IR: Infrared, NIR: Near Infrared, MRI: Magnetic Resonance Imaging.



Fig. 1. Electromagnetic radiation spectrum

and are often used for irradiation and plant breeding applications. Only a few studies have focused on agricultural product authenticity and geographical origin traceability by using gamma-ray and X-ray imaging techniques. In one study, gamma-ray analysis was conducted to distinguish beef products (Saito et al. 2008); the results showed that



Fig. 2. Scanning system for hyperspectral imaging A: Indoor scanning system: includes fixed trestles, removable sample platform, two lights, line cable, computer, and spectrographs; B: Outdoor scanning system: includes diesel generators, electric head, computer, and spectrographs.

Japanese black cattle beef and beef from the USA could be grouped separately using this method.

2. Ultraviolet imaging

Ultraviolet (UV) spans from 10 to 400 nm, with a broad wavelength range. As UV has shorter wavelengths than visible light, it can be easily scattered by the surface topology of materials, which helps to resolve or detect even smaller and finer characteristics. According to the different principles of performance, UV imaging can be divided into reflected UV imaging (used in astronomy and forensic science) and fluorescent UV imaging (used in molecular biology).

Although the application of UV imaging in the area of agricultural and food products is increasing (Al-Mallahi et al. 2010, Momin et al. 2011), UV imaging remains an emerging field of research, given the lack of suitable costeffective UV detectors currently available for food products. UV imaging offers remarkable potential for application in food agricultural product safety and authenticity. Therefore, along with the evolution of new-generation UV detectors and cameras, considerable research will focus on the application of UV imaging in the food industry (Manickavasagan et al. 2014).

3. Hyperspectral imaging

Hyperspectral imaging (HSI), also known as imaging spectroscopy, is an emerging technology originally developed for remote sensing, and which has many applications in resource management, agriculture, mineral exploration, and environmental monitoring.

As a new powerful analytical tool for nondestructive agricultural and food products analysis, HSI collects and processes information from across the electromagnetic spectrum, and then integrates conventional imaging and spectroscopy to attain both spatial and spectral information from an object. For each pixel in an image, a hyperspectral camera acquires the light intensity for numerous contiguous spectral bands. Every pixel in the image thus contains a continuous spectrum (in radiance or reflectance) that can be used to characterize objects in the scene with greater precision and detail. Push-broom HSI systems typically contain the following components: objective lens, spectrograph, camera, acquisition system, translation stage, illumination source, and computer (Fig. 2) (Zhao et al. 2010, 2012).

Spectral reflectance is the ratio of reflected radiant flux to incident radiant flux as a function of wavelength. For most materials, radiant flux at certain wavelengths is scattered or absorbed to different degrees, which causes reflectance to vary with wavelength. Pronounced downward deflections of spectral curves mark the wavelength ranges for which the material selectively absorbs incident radiant flux. These features are commonly called absorption bands. The overall shape of a spectral curve and the position and strength of absorption bands in many cases can be used to identify and discriminate different materials (http://www. microimages.com).

The HSI technique covers the wavelength from 400 to 2500 nm (Fig. 3), including visible light and reflected infrared (Randall B. Smith 2012, Butcher et al. 2016). The



Fig. 3. The best hyperspectral wavelength bands to study agriculture, in particular

wavelength is divided into three parts for plants (vegetation): (1) 400-700 nm, visible, chlorophyll absorption occurs in this area, usually used for leaf pigment measurement; (2) 700-1400 nm, near-infrared, reflectance due to spongy mesophyll is found in this area, usually used for cell structure detection; and (3) 1400-2500 nm, short-wave infrared, water absorption is included in this area, usually used for water content monitoring.

Zhao et al. (2010) used HSI to evaluate the potential of a portable spectral camera (400-1000 nm) for measuring cover crop chemical composition at different growth stages. They found that HSI could be used for evaluating agricultural resources, crops, and product quality. Zhao et al. (2012) also estimated the potential of soil organic carbon by conducting qualitative analysis using visible spectral graphs (400-1000 nm), near infrared spectral graphs (900-1700 nm), and a combination of both graphs. The results showed that these methods could be used to predict the soil organic carbon content.

Kamruzzaman et al. (2014) used HSI to conduct noninvasive authentication of the geographical origin of beef and pork by using the complete spectral range (380-1000 nm), and achieved an overall estimation accuracy of 95.0%.

Guo Z et al. (2013) conducted the geographical classification of apple based on HSI by using the wavelength from 400 nm to 1000 nm. The total classification rate had high accuracy of 89.86%.

In addition, Gao J, et al. (2013) used HIS (874-1734 nm) to discriminate the different geographical origins of *Jatropha curcas* L. seeds. The spectral and morphological features suggested a high discrimination rate of 93.75% for

the prediction of samples.

The HSI technology is a super nondestructive measurement method with wide applications in different areas. It can be utilized for assessing quality, especially for the geographical assessment of agricultural products in the future.

4. Fluorescence spectrometry

Fluorescence spectrometry, also known as a fluorescence fingerprint, is a rapid, simple and inexpensive method of measuring an object in solution based on its fluorescent properties. If some object is placed under irradiation, a beam of light (usually UV light) excites the electrons in the molecules of certain compounds and causes them to emit light that is typically, but not necessarily, vavisible light. Qualitative or quantitative analysis can be conducted by measuring both the excitation spectrum (i.e., light absorbed by the sample) and/or an emission spectrum (i.e., light emitted by the sample). Hashimoto et al. (2006) used fluorescence spectrometry to analyze the nutrimental components of tomato leaves; X-rays were used for excitation to obtain the X-ray fluorescence of fresh tomato leaves. The experimental results revealed that this method can be used to obtain nutrient information about the chemical elements and organic components. A high correlation coefficient (R=0.976) was obtained between the measured and predicted values of Ca content in tomato leaves.

In fluorescence spectroscopy, if the excitation beam has a varying wavelength, the fluorescence fingerprint will be a set of fluorescence spectra acquired at consecutive excitation wavelengths to create a three-dimensional

diagram. The pattern of this diagram contains abundant information about the constituents of a sample and is unique for each measured sample. By combining current information technologies and a large amount of data, different samples can be distinguished at a fine level (Sugiyama & Tsuta 2013). Nakamura et al. (2012) used this method to determine the geographic origin of mangoes. The misclassification rates of validated samples were 3.2-19.2% in 2010 and 7.7-13.6% in 2011. They suggested that the fluorescence fingerprint could be a practical method of determining the geographic origin of mangoes. Wavelengths from 260 to 290 nm for excitation and from 340 to 360 nm in emission were effective variables, and the fluorescence fingerprint is more suitable for skin than for pulp with lower misclassification rates. Nakamura et al. (2013) used the same method to determine the geographic origin of taros and obtained a misclassification rate of 9.4%. They suggested the application of this technology for predicting the buckwheat flour ratio in commercial dried buckwheat noodles, and for detecting mycotoxins in wheat and nutmeg.

The fluorescence fingerprint is a rapid, simple, accurate and cost-effective method of measuring nutrimental components and determining the geographic origin of agricultural products, and has the potential for wide-ranging applications.

5. Nuclear magnetic resonance imaging

Nuclear magnetic resonance (NMR) is based on the absorption and emission of energy in the radio frequency range from 60 to 1000 MHz of the electromagnetic spectrum (Mahajan et al. 2015). The principle of this technique is that many nuclei are included in a sample and all nuclei are electrically charged; when an object is placed in a strong magnetic field, the hydrogen nuclei spins become aligned parallel and anti-parallel to the field. The electromagnetic wave is applied to excite nuclear spins; after its removal, the signal strengths in orthogonal directions are measured. The measured signals are transformed using reconstruction algorithms to obtain high-quality images of the internal part of an object in two or three dimensions.

More attention is paid to the NMR method as a nondestructive technology and its potential for investigating the physical or biological properties of agricultural and food products. The use of NMR imaging to study food structure has been discussed using fish, meat (Han et al. 2014), milk, and emulsion samples (Haiduc et al. 2007, Bernewitz et al 2014, Ling et al. 2014). Most studies have focused on the water content of agricultural products, such as water distribution in soaked and cooked soybean seeds (Hong et al. 2009) or water absorbed by navy beans during cooking (Zhang & McCarthy 2013). In a study by Sekiyama et al. (2012), NMR spectroscopy and imaging technique were used to develop a new approach to identify the diseaseresistant cultivars of potato. They also conducted a study on apple juice by using the NMR technique in component profiling for product traceability.

6. Terahertz spectroscopy

Terahertz spectroscopy is another nondestructive and noninvasive monitoring technique that was recently recognized as being promising in the area of food quality and safety. Terahertz radiation corresponds to the range between microwave and infrared wavelengths, and consists of electromagnetic waves with wavelengths ranging from 1 mm to 100 µm (Zou & Zhao 2015). Unlike other conventional techniques, terahertz spectroscopy has significant advantages as it can pass through nonconductive materials and organic materials, such as wood, plastic, skin, paper, and clothing (Gowen et al. 2012). The development of THz time-domain spectroscopy (THz-TDS), which is based on determination of the THz pulse waveform and recovery of the frequency spectrum (Fitch & Osiander 2004) from the waveform by using Fourier transform, has promoted the usage of terahertz spectroscopy for various applications. THz imaging allows the direct measurement of absorption coefficients and has remarkable potential for the detection of pesticides and antibiotics in food powders by using THz-TDS. For example, some pesticides have an absorbance peak signature in the THz range, while food substance spectra appear to be the same. High absorbance of THz radiation by water enables its application for the quantification of moisture content in foods. Moreover, terahertz spectroscopy has applications in food quality monitoring, such as the detection of residue and foreign matter in foods, characterization of optical properties, and prediction of sugar and alcoholic contents (Gowen et al. 2012). Despite the remarkable information regarding terahertz imaging and the wide possibilities for its application, it still suffers from certain limitations, the most significant of which is its high cost, which is likely to be overcome by further technology development.

7. Other sensor techniques

In addition to the aforementioned nondestructive measurements using the electromagnetic spectrum, other sensor techniques are also used for assessing agricultural product authenticity and geography origin traceability. For example, the electronic nose and electronic tongue are adopted as gas and liquid sensor array systems for food quality control and composition assessment. The electronic nose focuses on volatile components that are released by samples, whereas the electronic tongue is often used for measuring the dissolved organic and inorganic components of food, usually liquid. Both cover a wide range of application, including dairy products, fish and meat products, fruits and vegetables, tea and coffee, as well as nonalcoholic and alcoholic beverages (Downey et al. 2016).

Challenges and prospects

The assessment of agricultural product authenticity and geography origin traceability may remain a very important issue relative to agricultural product quality and safety management. Previous studies have developed successful methods, such as stable isotopic analysis, compositional analysis, sensor technology, and spectroscopic techniques. Each method has its own advantages and disadvantages. For example, the advantages of isotopic analysis are good accuracy, high sensitivity, and wide application, whereas its disadvantages include high cost and a lengthy process. Nondestructive measurement has remarkable potential in the assessment of food authenticity and geographic origin traceability, due to a more rapid determination and reliable assessment as compared with traditional manual measurements or other instrumental measurements. Moreover, the advances made in optics have facilitated the rapid development of optical spectroscopic techniques in recent years.

Nondestructive measurement by using the electromagnetic spectrum for identifying agricultural product authenticity and geographic origin traceability is rapidly increasing along with technological advancement and lower equipment costs. In addition, the benefits of imaging spectrometry techniques over chemical, biochemical or molecular analysis for agricultural product applications are gradually being recognized and accepted, given their speed of detection, minimal or no requirement for sample preparation, possible simultaneously measurement of multiple attributes, and accurate detection. However, when measurements are performed using equipment, sensors or other devices, the accuracy of the data obtained must be improved for practical application.

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