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Changes in the physicochemical characteristics, including flavour components and Maillard reaction products, of non-centrifugal cane brown sugar during storage



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ABSTRACT

Changes in the quality attributes of non-centrifugal cane brown sugar represented by physicochemical characteristics as well as flavour components and Maillard reaction products (MRPs) were monitored every 3 months over 1 year of storage. Stored cane brown sugar became darker, and its moisture content and water activity (a_w) increased during storage. Fructose and glucose levels decreased as non-enzymatic browning via the Maillard reaction occurred in the stored sample, and a similar trend was also discovered in acetic and acetic acids. Stored cane brown sugar lost its acidic and sulfuric odours (58.70–39.35% and 1.85–0.08%, respectively); subsequently, the nutty and roasted aroma increased from 26.52% to 38.59% due to the volatile MRPs. The browning rate of stored cane brown sugar was positively associated with the development of volatile MRPs (Pearson's coefficient = 0.860), whereas the amount of 3-deoxyglucosone, an intermediate product of the Maillard reaction, had a lower association with the brown colour due to its relatively slow degradation rate.

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1. Introduction

Cane brown sugar is a valuable nutritional product from the sugarcane industry. It is produced by dehydrating sugarcane juice without centrifugation, and has many different names across the country, for example, kokuto in Japan, panela in South America, and jaggery in South Asia and Africa. This cane brown sugar, also called non-centrifugal sugar, has various biological functions with beneficial effects on human health, including anticariogenic, anti-toxic-cytoprotective, anticarcinogenic, and antioxidant effects (Jaffé, 2012; Payet, Sing, & Smadja, 2005). Cane brown sugar, particularly Japanese kokuto, contains bioactive compounds, including phenolics and policosanols, which have antiatherosclerotic activities (Asikin, Chinen, Takara, & Wada, 2008; Inafuku et al., 2007; Okabe et al., 2009; Takara et al., 2003).

Food storage is commercially required for preserving food materials and products prior to use. Hence, changes, both desirable and undesirable, in various food quality attributes commonly occur during storage, including changes in physicochemical properties, flavour components, and sensory characteristics (Jensen, Oestdal,

Skibsted, Larsen, & Thybo, 2011; Tosun & Ustun, 2003). Cane brown sugar can be stored for 1–2 years and is used either as table sugar and a snack or as raw material for the production of confectionery, beverage, and bakery products in Japan. However, very little information is available regarding the physicochemical composition and flavour constituents of stored cane brown sugar.

Non-enzymatic browning via the Maillard reaction, which involves a complex between amino acids and reducing sugars, can occur during food processing and storage. It also has critical effects on food properties, including the nutritional value, colour, texture, and flavour of sugary products, coffee, and chocolate (Martins, Jongen, & van Boekel, 2001; Zhou, Guo, Liu, Liu, & Labuza, 2013). The flavour characteristics of Maillard reaction products (MRPs) may vary from a pleasant, flowery, and fragrant aroma to a burnt, pungent, nutty, and caramel-like odour, depending on the amino acid and sugar composition in the food system, as well as their reaction pathways (van Boekel, 2006; Wong, Aziz, & Mohamed, 2008). Besides their nutritional and organoleptic qualities, MRPs have important biological functions such as antioxidant and anti-inflammatory activities (Chen & Kitts, 2011). Moreover, 3-deoxyglucosone (3-DG), one of the intermediate products of the Maillard reaction that is derived from glucose through non-oxidative formation, is a precursor of 5-hydroxymethylfurfural generation, indicating the degree of browning in food systems (Tosun & Ustun, 2003; Usui et al., 2007).

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This study aimed to determine the physicochemical characteristics, including colour, moisture content, water activity, sugars, and organic acids, as well as the flavour components of cane brown sugar during 1 year of storage. The volatile MRPs and 3-DG contents of cane brown sugar during storage were also evaluated. Furthermore, the influence of the Maillard reaction on the browning rate of stored brown sugar was investigated.

2. Materials and methods

2.1. Standards and reagents

Standard sugars (sucrose, fructose, and glucose), citric acid, malic acid, and succinic acid were obtained from Wako Pure Chemical Industries (Osaka, Japan). Aconitic acid and acetic acid were purchased from Nacalai Tesque, Inc. (Kyoto, Japan), and lactic acid, from Kanto Chemical Co., Inc. (Tokyo, Japan). Chemicals used as standards for analysing flavour components were obtained from Tokyo Chemical Industry (Tokyo, Japan), Sigma–Aldrich (St. Louis, MO, USA), and Wako Pure Chemical Industries. 2,3-Diaminonaphthalene (DAN) and 2-(2,3,4-trihydroxybutyl)-benzo[g]quinoxaline (3-DGad) were purchased from Dojindo Molecular Technologies, Inc. (Kumamoto, Japan), and 3-deoxyglucosone (3-DG), from Toronto Research Chemical, Inc. (Toronto, Ontario, Canada). All other reagents were from Wako Pure Chemical Industries and were of analytical grade.

2.2. Samples

Brown sugar was obtained from a local brown sugar factory (Iriomotetougyou Co., Ltd., Okinawa, Japan) and was a new product of the 2009 production year. The hardened block of brown sugar was crushed into powder or to various sizes of pebbles (1–3 cm) by a crushing machine. The crushed brown sugar samples (20 kg each) were immediately packaged in linear low-density polyethylene-coated double-paperboard 90 gsm bags (85 × 42 × 8 cm³; thickness, 60 μm) and then sealed. The samples were stored at room temperature in a closed storage room for 0, 3, 6, 9, and 12 months (mo) without humidity control. The day-night temperature and humidity changes during storage can be seen in Fig. S1 (supplementary data). The annual average of storage temperature was 26.8 °C with 52.1% relative humidity. After storage, the brown sugar was milled into powder at 25,000 rpm for 3 min using a Wonder Blender WB-1 (Osaka Chemicals Co. Ltd., Osaka, Japan) to pass a screen of 1.7 mm. The powdered brown sugar samples were kept at –30 °C prior to analysis.

2.3. Colour analysis

The colour of cane brown sugar was determined in terms of the international unit of colour for sugary products as per the standard ICUMSA (International Commission for Uniform Methods of Sugar Analysis) GS1/3-7 protocol (ICUMSA, 2003). Briefly, 1 g of sample was dissolved in 100 ml distilled water, and then adjusted to pH 7.0 by adding NaOH solution. The mixture was filtered through Advantec No. 2 filter paper and a 0.45-μm membrane filter (Toyo Roshi Kaisha, Ltd., Tokyo, Japan), and its absorbance was measured at 420 nm using a Shimadzu UV-1800 spectrophotometer (Shimadzu Corp., Kyoto, Japan). The sample colour was calculated using the equation: ICUMSA colour unit or IU = (absorbance × 1000)/(b × C), where b is the cell path length used (cm) and C is the concentration of sugar solution (g/ml). The sugar analysis is described in Section 2.5. All assays were performed in triplicate.

2.4. Moisture content and a_w analyses

Moisture content was evaluated based on the weight loss of a 5-g sample during oven drying at 105 °C for 1 h under atmospheric pressure and was expressed as percentage (%) of the total weight. A_w was measured with 7.5 g of sample using a Novasina IC-5000 AW-LAB water activity analyser (Novasina AG, Lachen, Switzerland). All assays were performed in triplicate.

2.5. Sugar composition analysis

The sugar composition of cane brown sugar was determined using a high-performance liquid chromatography (HPLC) method adapted from Xu, Niimi, and Han (2006) with slight modifications. Briefly, 0.3 g of sample was dissolved in 20 ml distilled water, and then filtered through a Millex-LH 0.45-μm membrane filter (Millipore Corp., Billerica, MA, USA). The columns used were Shodex SUGAR KS-801 and KS-802 columns (300 × 8.0 mm² internal diameter [i.d.]; 6-μm particle size; Showa Denko Co. Ltd., Tokyo, Japan) connected to a Shodex SUGAR KS-G (50 × 6.0 mm² i.d.) guard column. A TOSO-8010 HPLC system (Toso Co. Ltd., Tokyo, Japan) equipped with a refractive index detector model RI-8010 was used. The TOSO-CCPD pump was operated in isocratic mode with distilled water at a flow rate of 1 ml/min, and the CO-8010 oven was programed at a constant temperature of 60 °C. Samples and standards (30 μl) were injected through an AS-8010 auto sampler. The composition and concentration of sugars were calibrated by plotting peak area against concentration for the respective sugar standards, and expressed as grams per 100 g cane brown sugar. All assays were performed in triplicate.

2.6. Organic acid composition analysis

The organic acid composition was determined using an HPLC method adapted from Ji, Ji, Li, and Lu (2009) with slight modifications. Briefly, 1 g of sample was dissolved in 5 ml distilled water, centrifuged at 12,000 rpm for 5 min, and then filtered through an Advantec 0.45-μm membrane filter (Toyo Roshi Kaisha, Ltd.). Two Shim-pack SCR-102H columns (300 × 8 mm² i.d., Shimadzu Corp.) with a guard column (50 × 6 mm² i.d.) were maintained at 40 °C in a Shimadzu CTO-10AC oven. Two Shimadzu LC-10AD pumps were operated to stream the mobile phase containing 8.5 mM *p*-toluenesulfonic acid and post-column detection reagent containing 8.5 mM *p*-toluenesulfonic acid, 100 μM EDTA disodium salt, and 20 mM Bis-Tris buffer in the isocratic mode at a flow rate of 0.7 ml/min. The mobile phase and post-column detection solvent were streamed to a post-column reactor and mixed at a ratio of 1:1, prior to detection with a Shimadzu CDD-6A conductivity detector. The injection volume of samples and standards was 10 μl. The composition and concentration of organic acids were calibrated by plotting peak area against concentration for the respective acid standards, and expressed as milligrams per 100 g cane brown sugar. All assays were performed in triplicate.

2.7. Flavour component analysis

The flavour components of cane brown sugar were extracted using the solid-phase microextraction (SPME) technique and analysed using gas chromatography-flame ionisation detection/mass spectrophotometry (GC-FID/MS) (Poinot et al., 2007). Briefly, 3 g of sample was placed in a closed vial and heated in a water bath at 60 °C for 3 min. The volatile flavour components were then absorbed onto an SPME fiber containing divinylbenzene/carboxen/polydimethylsiloxane (Supelco Inc., Bellefonte, PA, USA) whilst heating for 20 min. The GC-FID analysis was performed using an Agilent 7890A GC system equipped with a fused silica capillary

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