Food Chemistry 168 (2015) 294–301

Contents lists available at ScienceDirect

Food Chemistry

journal homepage: www.elsevier.com/locate/foodchem

Effects of polishing, cooking, and storing on total arsenic and arsenic species concentrations in rice cultivated in Japan

Shigehiro Naito [a,*], Eri Matsumoto [b], Kumiko Shindoh [a], Tsutomu Nishimura [b]

[a] National Food Research Institute, National Agriculture and Food Research Organization, 2-1-12 Kannondai, Tsukuba, Ibaraki 305-8642, Japan
[b] Japan Food Research Laboratories, 6-11-10 Nagayama, Tama-shi, Tokyo 206-0025, Japan

ARTICLE INFO

Article history:
Received 13 February 2014
Received in revised form 5 June 2014
Accepted 9 July 2014
Available online 18 July 2014

Keywords:
Rice
Total arsenic
Inorganic arsenic
Polishing
Washing
Cooking
Storage

ABSTRACT

The effects of polishing, cooking, and storing on total arsenic (As) and As species concentrations in rice were studied adopting typical Japanese conditions. Total and inorganic As levels in three white rice samples polished by removing 10% of bran by weight were reduced to 61–66% and 51–70% of those in brown rice. The As levels in the white rice after three washings with deionized water were reduced to 81–84% and 71–83% of those in raw rice. Rinse-free rice, which requires no washing before cooking because bran remaining on the surface of the rice was removed previously, yielded an effect similar to that of reducing As in rice by washing. Low-volume cooking (water:rice 1.4–2.0:1) rice to dryness did not remove As. The As content of brown rice stored in grain form for one year was stable.

1. Introduction

Rice is a major source of inorganic arsenic (iAs), particularly in Asia and other countries where it is a staple food (Benford et al., 2011). The International Agency for Research on Cancer (2012) has classified iAs (arsenite As(III) and arsenate As(V)) as human carcinogens. The predominant As species in rice are As(III), As(V), and dimethylarsinic acid (DMA) (Heitkemper, Vela, Stewart, & Westphal, 2001; Huang, Fecher, Ilgen, Hu, & Yang, 2012; Meharg et al., 2008; Narukawa, Hioki, & Chiba, 2011; Nishimura et al., 2010; Williams et al., 2005; Zavala, Gerads, Gürleyük, & Duxbury, 2008; Zhu et al., 2008). Some literature also includes monomethylarsonic acid (MMA) (D'Amato, Forte, & Caroli, 2004; Pizarro, Gómez, Palacios, & Cámara, 2003), arsenobetaine (AsB) (Mihucz et al., 2007; Pizarro et al., 2003), or arsenocholine (AsC) (Mihucz et al., 2007).

The level of iAs in consumed rice depends on environmental conditions (As in soil and water), agricultural conditions (materials, cultivars, and control of irrigation water), and rice-processing and preparation methods (Benford et al., 2011). Reports of iAs concentrations in raw rice for risk assessment have been increasing, thus increasing the need for determining iAs in rice in order to assess the risk to humans (Carbonell-Barrachina et al., 2012; Juhasz et al., 2006; Laparra, Vélez, Barberá, Farré, & Montoro, 2005; Rintala, Ekholm, Koivisto, Peltonen, & Venäläinen, 2013; Schoof et al., 1998, 1999; Signes, Mitra, Burló, & Carbonell-Barrachina, 2008; Smith et al., 2006; Torres-Escribano, Leal, Vélez, & Montoro, 2008; Williams et al., 2005). However, few reports have focused on the effects of rice-processing and preparation methods on iAs content (Ackerman et al., 2005; Juhasz et al., 2006; Laparra et al., 2005; Mihucz et al., 2007; Narukawa et al., 2011; Raab, Baskaran, Feldmann, & Meharg, 2009; Signes et al., 2008; Smith et al., 2006; Sun et al., 2008; Torres-Escribano et al., 2008).

The Codex Committee on Contaminants in Food (CCCF), whose agenda included discussion of arsenic (As) in rice in the 1990s but discontinued it in 1999, has resumed this discussion since the Joint FAO/WHO Expert Committee on Food Additives (JECFA) re-evaluated the health risk of As exposure in 2010. CCCF has asked member countries, especially rice-producing countries, to submit

Abbreviations: As, arsenic; As(III), arsenite; As(V), arsenate; AsB, arsenobetaine; AsC, arsenocholine; CCCF, Codex Committee on Contaminants in Food; CRM, certified reference material; DMA, dimethylarsinic acid; DP%, degree of polishing; FAPAS, food analysis performance assessment scheme; HPLC, high-performance liquid chromatography; IARC, International Agency for Research on Cancer; iAs, inorganic arsenic; ICP-MS, inductively coupled plasma-mass spectroscopy; JECFA, Joint FAO/WHO Expert Committee on Food Additives; LOD, limit of detection; LOQ, limit of quantitation; MAFF, Ministry of Agriculture, Forestry and Fisheries of Japan; MMA, monomethylarsonic acid; NMIJ, National Metrology Institute of Japan; RSD_i, intermediate relative standard deviation; RSD_r, repeatability relative standard deviation; SD, standard deviation; tAs, total arsenic.

* Corresponding author.
E-mail address: naito@affrc.go.jp (S. Naito).

http://dx.doi.org/10.1016/j.foodchem.2014.07.060
0308-8146/

relevant data and information regarding As contamination in rice. Specifically, there is an urgent need to accumulate data on iAs in rice. Although Japan is a major rice-producing country, only a few studies have focused on total As and iAs concentrations in rice cultivated in Japan (Arao, Kawasaki, Baba, Mori, & Matsumoto, 2009; Kuramata, Abe, Matsumoto, & Ishikawa, 2011; Nagaoka, Nishimura, Matsuda, & Maitani, 2008; Narukawa et al., 2011; Nishimura et al., 2010).

In the present study, we investigate the effects of the following rice-processing and preparation treatments on total As and As species concentrations in rice by adopting a main rice cultivar (Koshihikari; (*Oriza sativa* L.) and a typical condition in Japan: polishing, processing rinse-free white rice, washing and low-volume cooking (water:rice 1.4–2.0:1), and storing rice in grain form for one year.

2. Materials and methods

The study design for treatment is presented in Table 1. We studied polishing and cooking as main treatments that cover most rice processing from harvest to table. In addition, we studied rinse-free rice, washing, and storage as supplement treatments.

Throughout the study, two analytical samples from each experiment were analysed. The experiments of individual treatments were performed in triplicate or in five replicates. Deionized water used throughout this study was purified in a Milli-Q system (Merck Millipore Corporation, Tokyo, Japan).

2.1. Samples

For selecting representative rice samples in Japan, we first chose the rice cultivar Koshihikari (*O. sativa* L.) because its share of total production is 40% and the shares of cultivars other than Koshihikari do not exceed 10% (Ministry of Agriculture, Forestry and Fisheries of Japan (MAFF), 2010). Furthermore, there were few genotypic differences in the level of either total As or iAs in the grains of 10 major Japanese rice cultivars including Koshihikari (Kuramata et al., 2011). Next, we determined the target total As levels (0.1 mg/kg, 0.3 mg/kg, and 0.5 mg/kg) in brown rice by reference to reports that (1) total As concentrations in 17 rice samples (brown rice, white rice, glutinous rice, and nonglutinous rice) produced in Japan ranged from 0.10 to 0.54 mg/kg (Nishimura et al., 2010) and (2) total As concentrations in brown rice from the major rice cultivars produced in Japan ranged from 0.04 to 0.33 mg/kg, with a mean of 0.16 mg/kg (n = 199) (MAFF, 2006). This MAFF survey reflected regional variations rather than genotypic variations (Kuramata et al., 2011). We thus collected four brown rice samples (ID = 1–4) with different total As levels from different regions in Japan (Table 1), and used sample 2 or sample 3 as a sample with a representative total As level in domestic rice. Sample 1 (180 kg = 30 kg/bag × 6 bags) was purchased from one farmer, and samples 2 (1050 kg = 30 kg/bag × 35 bags) and 3 (180 kg = 30 kg/bag × 10 bags) were purchased from another farmer. Sample 4 (90 kg = 30 kg/bag × 3 bags) was procured from an agricultural research institute where water-saving cultivation was adopted. Each sample was mixed just after procurement and repacked in rice bags, which were additionally placed in polyethylene bags for storage until treatment. Two rice bags (60 kg) of each sample (samples 1, 2, and 4) were selected randomly and then sorted using a grain colour sorter (ES-01AM, Satake, Hiroshima, Japan) that sorts sound rice grain from unsound rice grain (e.g., broken, immature, or coloured grains). Japanese rice-milling factories are usually equipped with grain sorters. The percentages of sound rice grain were 98.8% for sample 1, 99.9% for sample 2, and 95.0% for sample 4. The sorted brown rice samples were used for polishing, washing, and cooking; unsorted brown rice was used for rinse-free rice (sample 2) and storage (sample 3). Rice samples in polyethylene bags were kept at 10 °C until treatment and at 2–8 °C from treatment to analysis.

2.2. Polishing

White rice with a degree of polishing (DP%) by weight of 90% or 95%, was prepared from brown rice samples 1, 2, and 4 using a household rice mill (RSKM5B, Satake) at Satake Corp. (Hiroshima, Japan). White rice with 95DP% (90DP%) was obtained from milling 5% (10%) of outer layers from brown rice. Brown rice (750 g) was polished in the mill per run, and four runs were performed in one experiment. Five-replicate experiments were conducted on different days. White rice of each experiment was sifted using a sieve with a 0.5 mm wire diameter and a 2.0 mm aperture. The remaining white rice grains were used for washing and cooking. White rice grains are generally sorted after polishing in Japanese rice-milling factories.

2.3. Rinse-free rice treatment

Rinse-free white rice requires no washing for cooking because bran remaining on the surface was removed during production. Methods of producing rinse-free white rice in Japan include the wash-and-dry method, the adsorbent method using rice bran or heated tapioca, and the brushing method. In this study, bran was removed by the adsorbent method using bran other than that of the sample.

We studied rinse-free treatment as a supplemental experiment because of the minimum sample amount (180 kg/run) required for this processing, and used brown rice sample 2 as a representative sample for this treatment.

Brown rice sample 2 was mixed, sorted, and polished using a rice mill (DCM-75, Toyo Rice Cleaning Machine) and then processed for rinse-free white rice ("BG" rinse-free rice equipment with a production capacity of 3 ton/h, Toyo Rice Cleaning Machine) at a rice-milling factory of Toyo Rice Cleaning Machine Co., Ltd. (Wakayama, Japan). Brown rice (180 kg) was polished, sorted, and processed in one experiment; and five-replicate experiments were conducted on different days. DP% of white rice at each run could not be obtained because the rice mill at the factory is not equipped with a balance. Bran removed from white rice during production of rinse-free white rice could not be collected because

Table 1
Samples used for each treatment.

Sample ID	Total As in brown rice (mg/kg)[a]	Moisture in brown rice (%)	Production area	Cultivation year	Main treatment		Supplement treatment		
					Polishing	Cooking	Rinse free	Washing	Storage
1	0.503	13.4	A	2012	○	○		○	
2	0.235	14.1	B	2011	○	○	○	○	
3	0.250	13.6	B	2012					○
4	0.048	12.5	C	2012	○	○			

[a] Expressed as mg/kg dry matter.