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Essential and non-essential/toxic elements in rice available in the Portuguese and Spanish markets

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ABSTRACT

Eighty-six rice samples, representing the most common rice brands sold in Portuguese and Spanish markets (either locally produced or imported), were analyzed for their content of 35 essential and non-essential/toxic elements using inductively coupled plasma-mass spectrometry (ICP-MS). The mean content of essential elements was: 1189 mg/kg (P), 746 mg/kg (K), 294 mg/kg (Mg), 84.7 mg/kg (Ca), 13.3 mg/kg (Zn), 8.8 mg/kg (Na), 8.3 mg/kg (Mn), 7.5 mg/kg (Fe), 1.9 mg/kg (Cu), 0.55 mg/kg (Mo), 0.18 mg/kg (Se) and 0.12 mg/kg (Co). However, significant differences exist between the different types of rice, with brown rice showing the highest content of most essential elements. The daily intake of essential elements resulting from the average Iberian (Portugal and Spain) per capita consumption of rice was calculated and its contribution to the Recommended Dietary Allowance (RDA) or Adequate Intake (AI) was estimated. Data showed that rice can be an important dietary source of P, Zn, Mn, Cu, Mo and Se but does not significantly contribute to the daily dietary intake of Ca, Na and Fe. The content of toxic elements was very low, indicating that rice can be regarded as a very safe food.

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1. Introduction

Rice (the seeds of *Oryza* spp., mainly *Oryza sativa*) is a staple food for almost two-thirds of the world's population (Borresen and Ryan, 2014). Rice plant is cultivated almost worldwide, but China, India and Bangladesh are the largest rice producing countries. In Europe, most rice production is located in Italy and Spain, with both countries accounting for about 80% of the total European production. Portugal only represents around 6% of the rice production in Europe (FAOSTAT, 2015a). Rice consumption in Portugal and Spain has been steadily growing over the last years, mainly because consumers are switching from high-protein diets to high-carbohydrate diets as a result of the decreased purchase power caused by the economic recession. Portugal has one of the highest per capita consumption of rice in Europe, with approximately 16 kg (milled equivalent) per year in 2011 (FAOSTAT, 2015b).

Several minerals are necessary for humans to normal functioning of the body. Although the adequate intake of most minerals is usually achieved, low intakes for extended periods of time can cause deficiency signs. Nowadays, severe cases of iron (Fe) and selenium (Se) deficiency still exist all over the world, and the dietary intake of other minerals such as calcium (Ca), magnesium (Mg), iodine (I) and zinc (Zn) may be inadequate in several countries (Li et al., 2013; Stein, 2010). The introduction of the Recommended Dietary Allowances (RDAs), defined as "the levels of intake of essential nutrients that, on the basis of scientific knowledge, are judged by the US Food and Nutrition Board to be adequate to meet the known nutrient needs of practically all healthy persons" was a very important milestone in human nutrition science. RDAs are established based on the estimated average requirements (EAR), taking into consideration the incomplete utilization of the ingested nutrient (bioavailability) and integrating a safety factor to account for inter-individual variability (FNB/IOM, 2006).

Rice is an important source of energy, vitamins, amino acids, minerals and other nutrients for humans. The mineral content of rice is highly influenced by the degree of polishing/milling. During this process, the bran (i.e., the germ/embryo and the aleurone/pericarp layers) is removed from the brown rice resulting in white rice grains (Hansen et al., 2012). Since the outer grain layers are richer in minerals than the inner core, a significantly lower mineral content is usually observed in the polished/milled grains (Kumar

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et al., 2010). Therefore, a wide variability between the different commercially available rice brands is expected to exist.

Due to its large consumption, the accurate determination of the elemental composition of rice is of utmost importance for estimating the population's dietary intake of nutrients and their exposure to toxic elements. Based on this background, the aim of the present work was to determine the content of a wide range of elements (essential and non-essential/toxic elements) in rice commercially available in the Portuguese and Spanish markets. The contribution of dietary rice consumption for the daily intake of essential and non-essential/toxic elements was also estimated.

2. Material and methods

2.1. Samples

Rice samples were obtained from several supermarkets located in Porto (Portugal) and Vigo (Spain) cities in May 2014. A total of 86 rice samples from different types were analyzed: white rice ($n = 56$), parboiled rice ($n = 13$), brown rice ($n = 11$) and wild rice ($n = 6$). Wild rice (*Zizania* spp.) was also included in this study due to its close similarity to *Oryza* spp. The brand, country of origin and type of rice for each sample is listed in Table S1 (Supplementary material).

2.2. Reagents

High purity HNO_3 (≥69% w/w, *Trace*SELECT®, Fluka, L'Isle d'Abeau Chesnes, France) and H_2O_2 (≥30% v/v, *Trace*SELECT®, Fluka, Seelze, Germany) were used as received. The internal standard solution was prepared by appropriate dilution of the AccuTrace™ (AccuStandard®, New Haven, CT) ICP-MS-200.8-IS-1 (100 μg/mL of Sc, Y, In, Tb and Bi) solution. Calibration standards were prepared from the 10 μg/mL multi-element ICP-MS standard solution (*Plasma*CAL SCP-33-MS, SCP Science, Baie-d'Urfé, Quebec, Canada), from the 100 μg/mL multi-ion chromatography standard solution (AccuSPEC, SCP Science, Baie-d'Urfé, Quebec, Canada) and from the 1000 μg/mL Hg standard solution (TraceCERT®, Sigma–Aldrich, Buchs, Switzerland). All solutions were prepared using ultrapure water (>18.2 MΩ cm at 25 °C) obtained with a Milli-Q RG (Millipore, Billerica, MA) water purification system.

2.3. Instrument and apparatus

A Milestone (Sorisole, Italy) MLS 1200 Mega high performance microwave digestion unit equipped with an HPR-1000/10 S rotor was used for closed vessel acid digestion of the samples. Inductively coupled plasma-mass spectrometry (ICP-MS) analyses were performed using an iCAP™ Q instrument (Thermo Fisher Scientific, Bremen, Germany), equipped with a MicroMist nebulizer, a baffled cyclonic spray chamber (peltier-cooled), a standard quartz torch and a two-cone design (sample and skimmer cones). High purity (99.9997%) argon (Gasin II, Leça da Palmeira, Portugal) was used as the plasma source. The ICP-MS instrument operational parameters were as follow: RF power—1550 W; plasma gas flow—14 L/min; auxiliary gas flow—0.8 L/min; nebulizer flow rate—0.95 L/min. The following elemental isotopes (*m*/*z* ratios) were monitored for analytical determinations: ^{7}Li, ^{9}Be, ^{23}Na, ^{24}Mg, ^{27}Al, ^{31}P, ^{39}K, ^{44}Ca, ^{47}Ti, ^{51}V, ^{52}Cr, ^{55}Mn, ^{57}Fe, ^{59}Co, ^{60}Ni, ^{65}Cu, ^{66}Zn, ^{75}As, ^{79}Br, ^{81}Br, ^{82}Se, ^{85}Rb, ^{88}Sr, ^{95}Mo, ^{107}Ag, ^{111}Cd, ^{118}Sn, ^{121}Sb, ^{137}Ba, ^{139}La, ^{140}Ce, ^{202}Hg, ^{205}Tl, ^{208}Pb, ^{232}Th and ^{238}U; ^{45}Sc, ^{89}Y, ^{115}In and ^{159}Tb were used as internal standards. The instrument was tuned daily for maximum signal sensitivity and stability as well as for low oxides and doubly charged ion formation using the Tune B iCAP Q solution (Thermo Fisher Scientific; 1 μg/L of Ba, Bi, Ce, Co, In, Li and U in 2% HNO_3 + 0.5% HCl). Limits of detection (LOD) were estimated from 10 measurements of the blank solution (2% v/v HNO_3) and are presented in Supplementary material (Table S2).

2.4. Samples pretreatment

Rice samples were homogenized by grinding in a blender and sieved through a nylon sieve of 1 mm mesh size before acid digestion. The high-pressure microwave digestion rotor accommodates 10 PTFE vessels. Of these 10 vessels, eight were samples, one was a blank, and one was the certified reference material IRMM 807 or BCR 679. This configuration was kept unchanged in each digestion procedure. Powdered samples (~500 mg) were weighed into the microwave oven PTFE vessels and 4 mL of HNO_3 (≥69% w/w) plus 1 mL of H_2O_2 (≥30% v/v) were added to each vessel. The mixture was heated using the following microwave program: 250 W for 1 min, 0 W for 2 min, 250 W for 5 min, 400 W for 5 min, and 600 W for 5 min. After cooling, samples solutions were diluted to 25 mL in decontaminated volumetric flasks with ultrapure water. All solutions were analyzed by ICP-MS. All samples were analyzed in triplicate. Results were expressed as mg/kg, on a dry weight (dw) basis.

2.5. Estimated daily intake of elements

The Estimated Daily Intake (EDI) of essential elements resulting from rice consumption was calculated based on the elemental content (C_{elements}; mg/kg dw basis) and the average per capita daily consumption of rice (DC_{rice}), using the following formula:

$$\text{EDI} = C_{\text{elements}} \times \text{DC}_{\text{rice}}$$

where EDI is expressed in mg/day and DC was assumed to be 35.5 g/person/day for Iberian (Portugal and Spain) consumers (FAOSTAT, 2015b). The obtained values where compared with RDA (Recommended Dietary Allowance) or AI (Adequate Intake) as suggested by FNB/IOM (2006). Cobalt was not considered since no RDA or AI have been established.

2.6. Provisional tolerable weekly intake

The Provisional Tolerable Weekly Intake (PTWI) of non-essential/toxic elements resulting from rice consumption was calculated based on the elemental content (C_{elements}; mg/kg dw basis), the average per capita weekly consumption of rice (WC_{rice}) and the individual's body weight (bw), using the following formula:

$$\text{PTWI} = \frac{C_{\text{elements}} \times \text{WC}_{\text{rice}}}{\text{bw}}$$

where bw was assumed to be 70 kg and WC was assumed to be 248.5, which was derived by multiplying the number of days in a week (i.e., 7) by 35.5 that is the daily per capita rice consumption for Iberian (Portugal and Spain) consumers (FAOSTAT, 2015b). The obtained values where compared with the PTWI values published by JECFA (2011).

2.7. Quality control

For quality control purposes, the certified reference material (CRM) IRMM 807 (rice flour, supplied by EC Institute for Reference Materials and Measurements, Geel, Belgium) and BCR 679 (white cabbage, supplied by EC Institute for Reference Materials and Measurements, Geel, Belgium) were analyzed under the same conditions as for the samples. The average recoveries obtained in the CRM analysis are presented in Table 1.

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